Prediction of the long-term leaching behaviour of a landfill containing predominantly inorganic waste

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PREFACE

The International Waste Working Group (IWWG) was established in 2002, following a world-wide demand, to serve as a forum for the scientific and professional community.

The aim of the IWWG is to provide an intellectual platform to encourage and support economical and ecological waste management, and to promote scientific advancement in the field. This aim is being accomplished by learning from the past and by analyzing the present, with a view to developing new ideas and visions for the future.

The objectives of the IWWG are pursued mainly by means of: collecting, developing and disseminating new results and ideas based on Research and Development; promoting discussion on strategic matters, providing and organizing education in waste management and transferring knowledge into practical applications.

To achieve these objectives, the IWWG publishes an international journal (*Waste Management*, Elsevier Publisher), organizes Symposia, Conferences and specialized Workshops, establishes specific Task Groups aimed at discussing the main aspects of waste management and technology, as well as identifying common positions to be proposed subsequently to regulators, decision makers and operators.

In keeping with the aims described in previous paragraphs, the IWWG has opted to publish a series of reference books focusing on the aspects of current interest in the field of waste management and technology.

Contributions to these books originate from relevant papers presented during events organized or promoted by IWWG (in particular the Sardinia Symposia) and from original contributions to the books. The overall intent is to make this wealth of information available to the waste management community in a concise and organized fashion.

The editors of the monographic volumes are internationals experts, generally members of the IWWG Managing Board or Scientific Advisory Panel. Editors perform a variety of tasks including selecting and organizing papers, standardizing the texts and eventually making constructive suggestions when the original manuscript undergoes significant changes. The responsibility for the technical content of the book lies with the individual Authors.

I wish to thank the Editors and all Contributors as well as Roberta Gadia, Tiziana Lai and Paola Pizzardini, for their efforts and support; I trust that the monographic book series and in particular the volume on hand will make a positive contribution towards creating a better understanding of the numerous aspects of waste management and supporting the procedure for making environmentally-safe and economically effective decisions.

Rainer Stegmann, IWWG President

The project for this monograph started a few years ago in the wake of the activities carried out by the Working Group on Sustainable Landfilling, at that time chaired by Raffaello Cossu.

Selected papers from the Proceedings of different Sardinia Symposia have been grouped in order to provide a well-organized and useful volume.

The editing of the work took longer than anticipate, which, combined with the dreadful daily routines we are all affected by, determined a delay in the completion of the final product.

We hope that a widespread interest in the publicaton by the audience can make up for the above delay, for which we sincerely apologize, especially with the authors.

R Cossu
H van der Sloot

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Introduction

Although more waste materials are recycled or re-used nowadays, landfilling of waste materials is still necessary in the future. The necessary everlasting aftercare of landfills raises concern. In the Netherlands, a Sustainable Landfill Foundation was set up in order to stimulate the development of new landfill technologies to minimize impact and thus reduce or eliminate the need for long-term aftercare. A five-year research program was started in order to develop and evaluate four sustainable landfill techniques (predominantly inorganic waste, organic waste, bioreactor and stabilised waste). The complete results of the project can be found in (Mathlener et al., 2006; van Zomeren and van der Sloot, 2006). The project aims at creating a biogeochemical equilibrium between a landfill and the environment within a period of 30 years, in order to reduce the long-term risk and the aftercare needs. This can be accomplished by constructing a landfill that has chemical properties that will minimize leaching of contaminants (e.g. neutral pH, slightly reduced conditions, low organic matter content and relatively low salt loads).

Waste acceptance criteria such as specified in the EU landfill directive (1999) are based on test results from individual waste materials. However, it is unclear whether these results have any significance with regard to the final behaviour and emissions of a complete landfill. In earlier work (van der Sloot et al., 2001) laboratory tests were performed with relatively small additions (up to 10%) of contaminated (or alkaline) waste materials to a stable (neutral pH, low organic carbon content) waste material (mix of sludge, soil remediation sludge and construction and demolition waste). We have shown that relatively small additions (up to 10%) of contaminated waste materials do not significantly disturb the leaching behaviour of the total waste mix. Key controlling factors for this predominantly inorganic waste disposal are controls on the individual waste dissolved organic carbon levels, mobile inorganic (e.g. Chloride and sulphate) and water-soluble organic contaminant levels.

The long-term leaching behaviour of a landfill is strongly affected by the macro-chemistry, which is currently largely ignored in monitoring of percolate water. At the start of the project additional parameters have been identified that needed to be measured to enable chemical speciation modelling in order to understand the chemical processes in the landfill leading to a release of contaminants. A major challenge is to develop means to predict the long-term leachate quality based on geochemical reactive transport modelling taking into account the geochemistry as well as preferential flow aspects, which are occurring in landfills. The anticipated outcome is a disposal practice through waste acceptance that will reduce the contaminant emissions to acceptable levels on the long-term. Predic-

tion of the long-term leaching behaviour of a landfill is particularly relevant for judging the need for aftercare.

In this study, the results from column leaching tests are described by geochemical reactive transport modelling. This approach will enable understanding of release processes under field conditions. Furthermore, preferential flow aspects will be addressed in the paper by comparison of the cumulative release of mobile species in lysimeter and field situations with the laboratory characterisation of the mixed waste entering the landfill. This gives an indication of the extent of preferential flow in the landfill compartment. This work provides a means of gaining more understanding and subsequently more control over the long-term release of inorganic contaminants and identification of possible long-term processes that can disturb the biogeochemical equilibrium of the landfill.

Materials and methods

Laboratory experiments, lysimeter and field measurements

In the framework of a Dutch national research project on sustainable landfill, laboratory experiments (percolation test – CEN/TS 14405, 2003; pH dependence test – CEN/TS 14429, 2003), lysimeter studies (1-1,5 m^3) and a 12,000 m^3 pilot demonstration project at landfill site Nauernasche Polder (the Netherlands) are carried out in conjunction with chemical speciation modelling and release modelling.

The filling of the pilot started in April 2000 and was completed in November 2001. The test cell is isolated from the rest of the landfill site by a HDPE membrane. Leachate is collected in the lower corner of the test cell and the amount of leachate pumped out of the test cell is measured. In the centre of the test cell a vertical drain (filled with coarse granular material) is installed to enhance the draining of rainwater and to minimise the contact of rainwater with the waste. The waste input to the pilot cell is controlled by more stringent acceptance criteria than currently required by regulation. Samples were taken from all waste streams deposited in the cell and the landfilled weight of each stream was recorded. Quality control measurements were made on-site by short leaching tests. Every batch of waste was leached at an L/S ratio of 2 L/kg for one hour. After centrifugation and filtration the sample was analysed for pH, electric conductivity, chloride and Dissolved Organic Carbon (DOC). From all waste samples collected an integrated waste mix was prepared by taking the waste mass per waste charge into account. This waste mix was used for the laboratory testing according to CEN/TS 14405 (coded Column test and Column test-ORG) and CEN/TS 14429 and for filling three lysimeters with a representative waste mixture (October 2001). Filling of the lysimeters was carried out as follows: Lysimeter 1 - Disposal of wastes in order of delivery as practiced at Nauerna (coded Lysimeter1-P and -R); Lysimeter 2: Encapsulation of more contaminated wastes in relatively low permeability wastes (soil cleaning residues; coded Lysimeter2-P and -R);Lysimeter 3: Disposal of wastes in order of delivery as practiced at Nauerna with addition of 5 w/w % of sewage sludge and car schredder waste each to increase

organic matter loading (coded Lysimeter3-P and -R). The studies at field, lysimeter and laboratory scale represent different time scales through the liquid to solid ratio, to which the waste was exposed.

Chemical analysis

The leachates and extracts from laboratory tests were analysed for major, minor and trace elements by ICP (Al, As, B, Ba, Ca, Cd, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Mo, Na, Ni, P, Pb, S, Sb, Se, Si, Sn, Sr, Tl, V, Zn). DOC and TIC were analysed by a Shimadzu TOC 5000a analyzer. Cl, F, ammonium and sulfate were analysed by ion-chromatography. PAH was analysed by HPLC, mineral oil and volatile organic halogenated compounds were analysed by GC-MS.

Geochemical speciation and release modelling

The geochemical modelling framework ORCHESTRA (Meeussen, 2003), using an extended MINTEQA2 database with thermodynamic constants for inorganic reactions, was coupled to a database/expert system (LeachXS)(van der Sloot et al., 2003a) containing the pH dependence leaching test data, the percolation test data, lysimeter and field leachate data for quick data retrieval, processing and data presentation. The role of Dissolved Organic Carbon (DOC) on metal mobilisation is addressed by applying the NICA-Donnan model (Kinniburgh et al., 1999). The measured availability for all major and minor elements was used as input for the model. A rapid batch method for the total humic acid (HA) concentrations in the waste mixture was used (van Zomeren and Comans, 2007) to estimate the relevant organic matter content. Dissolved HA concentrations were estimated to be 20% of the measured DOC content in the eluates. The amount of amorphous and crystalline iron (hydr)oxides in the waste mixture was estimated by a dithionite extraction described in Kostka and Luther III (Kostka and Luther III, 1994). The amount of amorphous aluminum (hydr)oxides were estimated by an oxalate extraction according to Blakemore (Blakemore et al., 1987). The extracted amounts were summed and used as an estimate for hydrous ferric oxides (HFO) in the model.

The time-dependent source term for the predominantly inorganic waste landfill is quantified and used for predictions of long-term leachate quality. Environmental impact modelling using the model developed for the ANNEX II of the EU Landfill Directive (1999; Hjelmar et al., 2001) allows a verification of performance of the new landfill concept against criteria.

The measured concentrations as a function of L/S ratio as determined with a column test (NEN 7343), were predicted with ORCHESTRA. Information on possible relevant mineral phases was derived from modelling the pH dependent leaching behaviour. Parameters for HA, FA and HFO were measured as described above, porosity was estimated to be 30%. The density of the material was 1800 kg/m^3. The model predicts the concentration, cumulative release and partitioning of major, minor and trace elements as a function of L/S (linked to a time scale) using one set of element availabilities, selected minerals, sorption parameters for clay, Fe-oxides and organic matter and flow characteristics in terms of flow velocity and ratio between mobile and stagnant zones.

Results and discussion

Leaching behaviour of contaminants at laboratory, lysimeter and pilot scale

The emissions of As, Cd, Cr, Ni, Pb and Zn are given in Figure 1 and Figure 2. In general, the emissions from all the tests are within a relatively narrow bandwidth; this also applies to the measured concentrations in solution. This means that the tests done on the three scales give a consistent view, and that in these cases the emissions measured in the column test are a good indication of emissions over the long-term. This evaluation will compare the emission from the column test, the lysimeter experiment containing waste such as that landfilled at Nauerna (Lysimeter 1) and the pilot project (Pilot). These three tests are the most similar. In addition, the release of pollutants from the lysimeter-2 mixture (more polluted streams packed into poorly permeable materials) gives comparable results in many cases. When extra organic matter is added, the results can deviate due to the different composition and can therefore not be directly compared with the other experiments. These results can however be compared to the emissions from the column test on this mixture (Column test-ORG).

The pilot project data for As lies on the higher side of the concentration range. The reducing conditions in the pilot project can reduce As(V) to As(III). This is a more mobile component than As(V). It should be noted that many measurements from the column test and lysimeters are approximately around the limit of detection for As. The emissions are consistent for the experiments on all the different scales. It is expected that the As emissions will not exceed the standard (LFD, inert) in the long-term.

The Cd concentrations in the first and second fractions from the column tests are markedly high. Thus the cumulative emission from the column test deviates greatly compared to the other experiments. The other concentrations are very low and often smaller than the limit of detection. This deviation could possibly be associated with $CdCl_2$ complexes. These complexes can increase the solubility of Cd. In Figure 1 it can be seen that the Cd concentrations in the first and second fractions of the column test are also relatively high in comparison with the other measurements. The pores are easily accessible and the concentration can become high because the column test can be effectively leached during the experiment (with respect to the preferential flow in the pilot project and lysimeters). When the first fraction of the column test is not taken into account, then the Cd emission is not critical. The results of the pilot project and the lysimeters also indicate that Cd is not critical with respect to the applicable legislation. Therefore, it can be concluded that Cd is not critical.

Cr emissions are not critical with respect to the LFD. The emissions also give a very consistent picture. The measured concentrations range from around the limit of detection to a maximum of a factor of 10 above.

Initially the Ni decreases slowly in the column test, and later more rapidly. It seems that the cumulative leaching could become critical when the waste mix contains too much organic matter; Ni complexes strongly with DOC. The leaching of Ni is not critical in the normal mix. The concentrations in the pilot project and the lysimeters also seem to decrease slightly

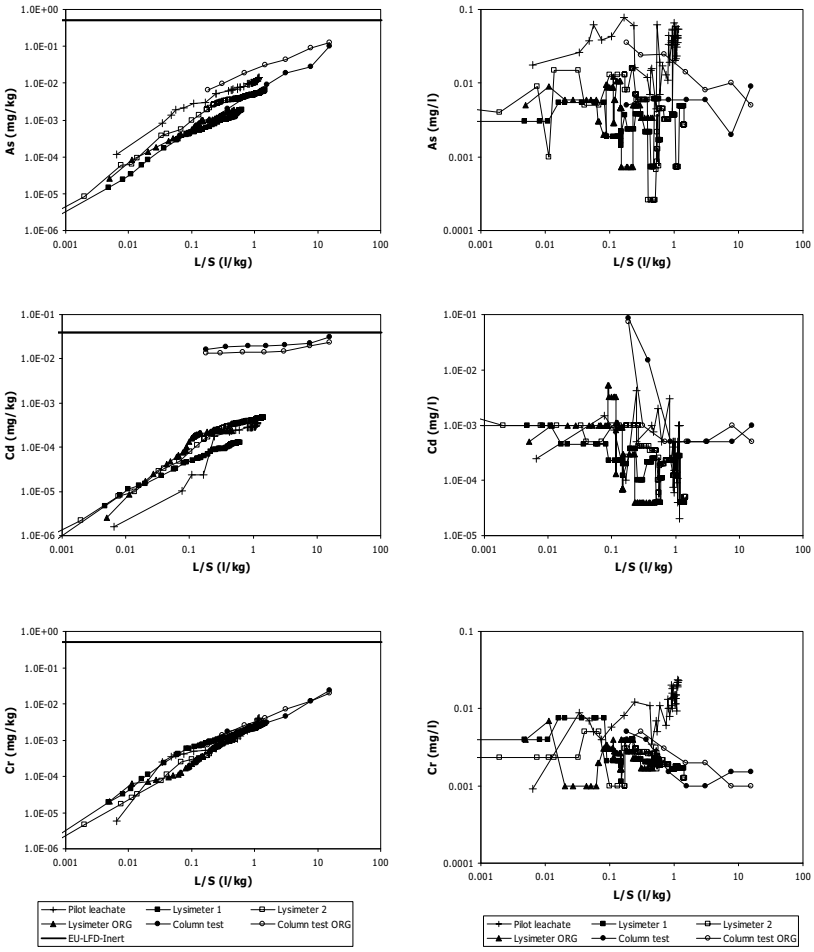
at higher L/S values. Further measurements would be able to show whether this trend continues.

The Pb concentrations are somewhat higher in the first fraction of the column tests than the other measurements. This also causes the emission curve to shift higher. The emissions are solubility-controlled in all the experiments. The pilot project also shows that depletion may now be taking place, as shown by a flattening of the emission curve. When the results of the column experiments are compared with the standards sets by the LFD, it is apparent that the emissions are approximately a factor of 2 under the standard. It is expected that the Pb emissions will also not be critical in the long-term, on the basis of the results from the lysimeter experiments and the pilot project.

The leaching of Zn shows relatively large differences between the different experiments. The pilot project shows the lowest emission; the column tests have the highest emissions. The leaching of Zn is not critical with respect to the European Landfill Directive for inert waste. The differences in leaching are too large to be explained by preferential flow in the pilot project and the lysimeters. Zn can form complexes with dissolved organic matter. It is possible that organically complexed Zn plays a greater role in the column test than in the other experiments. However, it is not yet clear how important this mechanism is in the experiments.

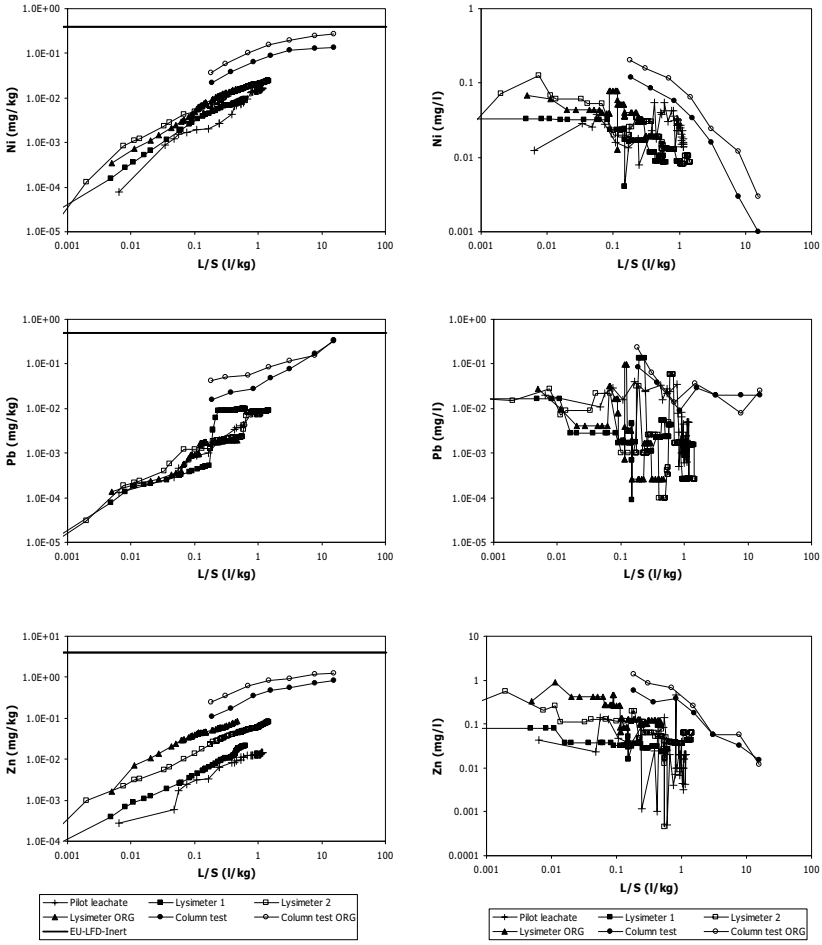
Leaching behaviour of redox sensitive elements at laboratory, lysimeter and pilot scale

As shown above and in earlier work (van der Sloot et al., 2003b), there is in general a very good agreement between the leaching behaviour of the constituted waste mixture and the leachate as obtained from the full-scale pilot experiment. This indicates that in spite of an apparently very heterogeneous mix of materials, leaching is governed by well-defined solubility controls for many constituents. However, widely different redox potentials have been measured in the laboratory, lysimeters and the pilot experiment (range roughly from -300 mV to +300 mV) as can be seen in Figure 3. The pilot experiment leachate has a relatively constant redox potential of about -200 mV, whereas the lysimeter experiments start with redox potential within the same range. After about 1.5 years after the start of the experiment, the redox potential in the lysimeter increases to more or less stable values of roughly +200 mV. Presumably, the waste material is oxidised by the influence of direct exposure to the atmosphere. The laboratory column leaching tests exhibit an increasing redox potential ranging from -100 mV at the start to values of about +250 mV at the end of the experiment (16 days). In this case the leachant is oxygen saturated water, which implies that the reducing capacity of the waste mix is limited. The effects of different redox conditions on the leaching behaviour of several relevant elements are discussed below.



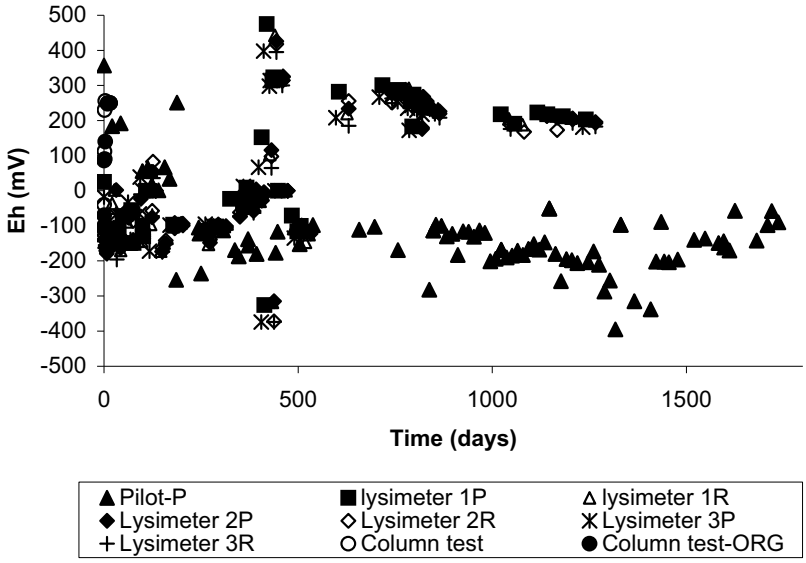
[Figure 1] Emissions of As, Cd, and Cr as a function of the L/S ratio in laboratory measurements (column test), lysimeter experiment and pilot project. The addition ORG is for the waste mixture with 10% additional organic waste. The solid black horizontal line represents the emission limit for inert waste.

The observed differences in redox potentials in the performed experiments lead to differences in the release of elements like Cu, Fe and Mn (Figure 4). The release of earlier presented data at different scales of testing (van der Sloot et al., 2003b) is still consistent, as an example the leaching of SO_4 is also presented in Figure 4. It can be seen that the SO_4 data from different scales of testing show a very consistent pattern and a close match of the cumulative release. The cumulative release of Cu is much lower in the pilot experiment compared to the other test types (lysimeter and column leaching tests). Fe and Mn show a relatively high leaching in



[Figure 2] Emissions of Ni, Pb and Zn as a function of the L/S ratio in laboratory measurements (column test), lysimeter experiment and pilot project. The addition ORG is for the waste mixture with 10% additional organic waste. The solid black horizontal line represents the emission limit for inert waste.

the pilot experiments, this release is in line with the laboratory column test results but much higher than the lysimeter experiments. The reason for these differences is probably the difference in redox potential in these leaching tests. In general, the redox potential in the leachates from the pilot experiment is around -100 to -200 mV. As the leachate collection system is not sealed, the redox potential in the pilot cell itself may actually be lower. The redox potential in the lysimeter experiments is generally about +200 mV. A low redox potential under field situations may reduce Cu(II) to Cu(I). The affinity of humic substances for metal complexation is



[Figure 3] Redox potential measurements (uncorrected measurements) as a function of time in laboratory (column test), lysimeter and pilot scale experiment with predominantly inorganic waste.

lower for Cu(I) species. Moreover, the formation of relatively insoluble copper sulphides might be a dominant process resulting in lower dissolved Cu concentrations.

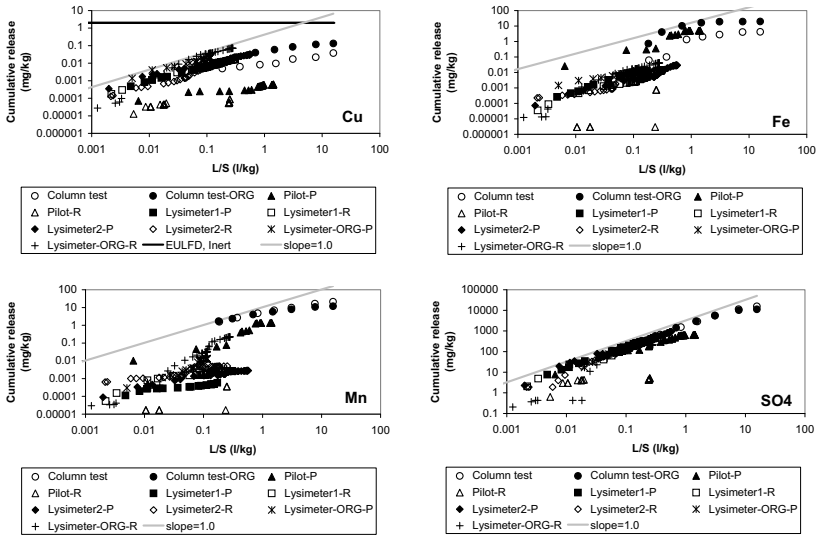
Fe is normally present as Fe(III) and can easily be reduced to Fe(II) in a reducing environment. The solubility of Fe(II) is much higher (e.g. (Jansen et al., 2003)), resulting in an enhanced leaching of total Fe. The column test results also show a relatively high Fe leaching, this might also be due to a reducing environment in several fractions during the column leaching test. The extent of reduction during a leaching test depends on the reducing properties of a material, which can be measured with a Cerium titration method (NEN 7348).

The leaching of Mn in the pilot experiments and the column experiments is higher than the results for the lysimeters. Under reducing conditions like in the pilot experiment and (to a lesser extent) the column test, the soluble Mn(II) species will be dominant. This species can easily be oxidised to Mn(III) which forms solid oxides (Pourbaix, 1966).

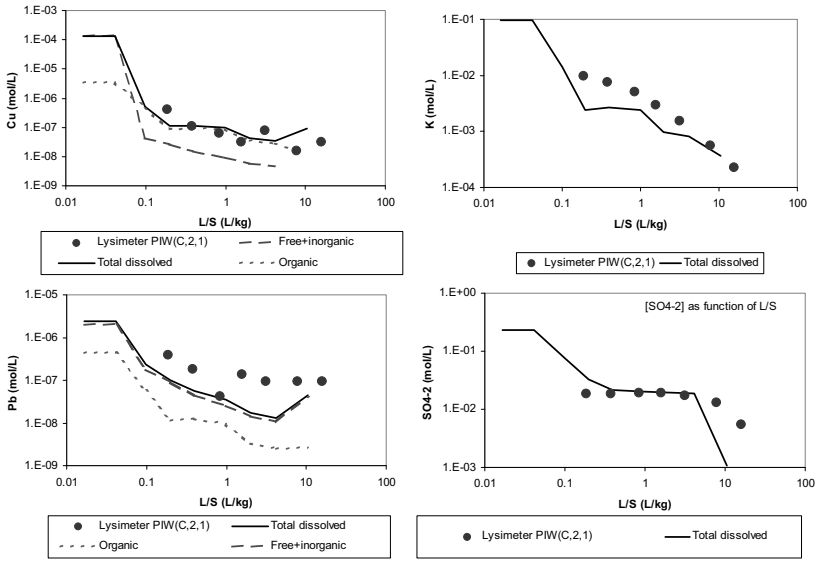
The leaching of Fe and Mn in the column test and the pilot experiment show a consistent behaviour, the leaching of these elements in the lysimeter experiments is significantly lower. However, Cu leaching in the column test is consistent with the lysimeter experiments. In this case the Cu leaching in the pilot experiment is significantly lower. These differences can possibly be explained on the basis of the different reducing conditions in the test methods. The redox potential in the lysimeter percolate water

is generally relatively high with respect to the standard redox potentials of the Cu, Fe and Mn reduction reactions (Stumm and Morgan, 1981). The eluates collected in the column test have a slightly lower redox potential, possibly low enough to reduce Fe and Mn but not low enough for reduction of Cu. Finally, the relatively low redox potentials in the percolate water from the pilot experiment are presumably low enough to also reduce Cu. Preliminary calculations with predominance diagrams also indicate that Cu(I) species are likely to be present under these conditions.

Geochemical release modelling under varying environmental conditions
Figure 5 shows the measured and predicted concentration from the waste mixture as a function of the L/S ratio. The model predictions start at an L/S ratio of 0.02 up to 10, the solid lines represent the predicted total dissolved concentrations. The dotted lines for Cu and Pb represent the organically complexed metal (green line) and the free + inorganically complexed metal (pink line) respectively.
The model generally predicts the release of Cu, K, Pb and SO_4^{2-} very well. This implies that the chemical processes leading to release are understood. The release of Cu is described well as can be seen from the predicted line for total dissolved Cu. It is known that Cu complexes strongly to natural organic matter (Buffle, 1988), the model prediction also shows that Cu is predominantly complexed to organic carbon. It is interesting to note that the model predicts a high Cu concentration at very low L/S ratio's and



[Figure 4] Cumulative release as a function of L/S for data from column leaching tests (Column test and Column test-ORG), lysimeter experiments (Lysimeter1-3) and a pilot-scale experiment (Pilot). The line with slope = 1 shows the slope of a solubility controlled release; The black line for Cu indicates the EU-LFD limit for inert waste.



[Figure 5] Measured and calculated emissions from waste mixture as a function of the L/S ratio. Data points represent measurements with the column test (NEN 7343), the lines represent the calculated species.

that Cu is predominantly in the free and inorganically complexed form. Additional experiments need to be performed to check whether this effect appears in column test measurements at these low L/S ratios.

Leaching of Pb is described adequately with a constant underestimation of the calculated concentration, which is up to one order of magnitude. Pb is predominantly leached in the free and inorganic form, about 10% is organically complexed.

The release of SO_4^{2-} is well described by the model. At high L/S ratio, the model predicts a stronger decrease in SO_4^{2-} leaching compared to the measurements. This effect is probably due to a large deviation in the calculated pH value at high L/S ratio's (0.1-0.3 units from L/S 0.1 to 7 versus about 1.5 units at L/S 10). We are currently focusing on possible causes of this deviation. The washout of dissolved organic matter appears to play a role in the deviation of the predicted pH.

The results for all other major, minor and trace elements (not shown here) are also very promising for prediction of the long-term release from this waste material. This approach and the level of detail in modelling transport processes is currently state of the art. However, several contaminants show deviations of over one order of magnitude and these deviations need to be sorted out in future work.

Leaching of K is controlled by the availability since this element is very mobile and not significantly retained in the matrix. The model prediction for this element is adequate and promising for future model calculations.

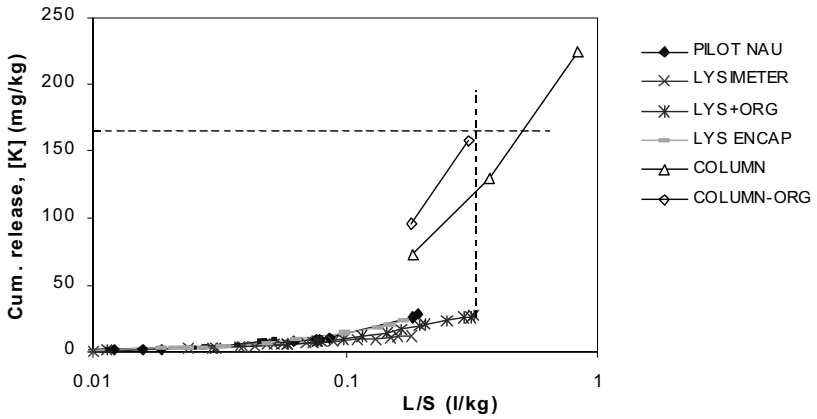
We have taken dual porosity into account in our calculations, assuming that in the column test 85% of the waste is effectively leached (15 % stagnant porewater or dry).

Preferential flow

The mass of all waste materials delivered to the pilot cell was recorded. This implies that by constituting the waste mix in proportion to the mass input a rather good balance of all constituents in the pilot cell is obtained. A comparison between the release of mobile constituents (Cl, Na, K) in lysimeters and field leachate with the column leaching test data obtained with upflow (minimal channelling) will allow conclusions to be drawn on the possible role of preferential flow. In Figure 6 the cumulative release of K is given as a function of L/S (l/kg). The release from lysimeters and from the pilot is lower than from the laboratory test and can be explained by preferential flow. From the comparison of release at the corresponding L/S a factor can be calculated. This has been done for Cl, K and Na as these parameters do not interact appreciably with the matrix. In Table 1 the release as derived from lysimeter and field data is given relative to release obtained for column leaching tests.

[Table 1] Calculated relative release for lysimeter and field to column data for mobile constituents. Calculations based on corresponding L/S values in the L/S range = 0.05 and 0.7.

Experiment	Na %	K %	Cl %
Nauerna Pilot	46	36	28
LYSIMETER	23	16	22
LYS + ORG	33	23	21
LYS ENCAP	38	33	36



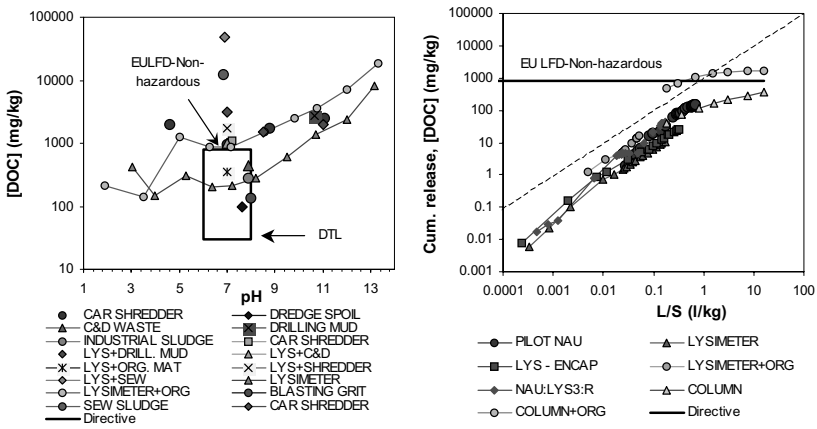
[Figure 6] Comparison of release of K derived from lysimeter (LYS) and field leachate data (PILOT) with results from up-flow column leaching tests (COLUMN).

Dissolved Organic Carbon

In Figure 7 the data for DOC from laboratory testing and leachate analysis in lysimeters and field are combined. The results from the pH dependence test illustrate a significant increase in DOC leachability after addition of organic rich materials (10 % w/w) to the integral waste mix. Data for individual wastes scatter much more widely. DOC increases with increasing pH, which is explained by the mobilisation of humic type substances as opposed to a more fulvic acid rich condition around neutral pH. The data obtained for the lysimeters, laboratory data and field leachate data form a consistent dataset with a good overlap between the different scales of testing. The L/S ratio in the pilot-scale and lysimeter experiments is determined by dividing the amount of leachate produced in a certain time period by the total amount of waste in the test cell or lysimeter. It is assumed that the test cell or lysimeter is totally filled with waste from the beginning of the experiment. This assumption is the most acceptable when comparing the column leaching test data with lysimeter and field data. Further monitoring in the field will be carried out to see if the match between lab and field is maintained and the decline in concentration as observed in lab testing also occurs in the field. It is striking to note the consistency in leaching of the integral waste mix composed from subsamples collected during filling of the pilot cell on both lab and lysimeter experiments and the results obtained from the pilot. This would imply that similar solubility controlling mechanisms are active at the different scales, thus increasing the potential for long-term prediction significantly.

Emissions of individual waste materials and their consecutive mixture

In Figure 8 the data for Pb from laboratory testing and leachate analysis in lysimeters and field are combined. The leaching behaviour of Pb



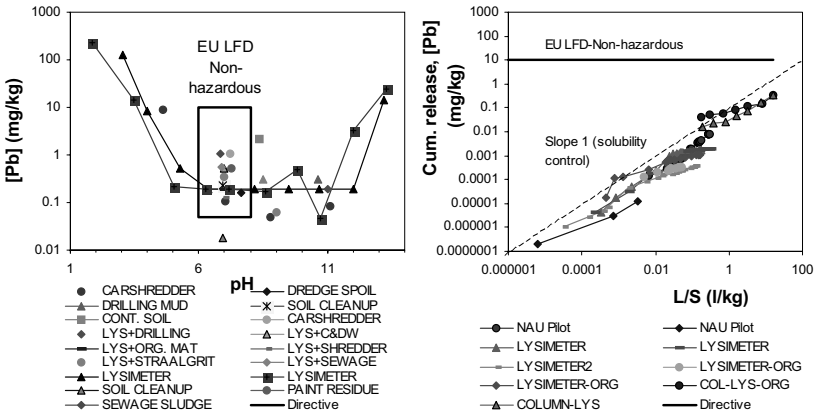
[Figure 7] Leaching of DOC as obtained from pH dependence test, percolation test, lysimeter and pilot cell leachate. Individual wastes are given in the plot on pH dependent leaching. Box represents most likely pH domain for the mixed waste. DTL = detection limit. EU Landfill limits for non-hazardous waste inserted for comparison.

from the waste mix is consistent with observations on other types of waste. The data for different wastes (individual measurements) scatter widely. The resultant integral waste mix, however, behaves very consistently and judging from the slope in the release L/S plot, its behaviour is controlled by solubility. The end point of the column test at L/S = 10 matches very well with the corresponding leached amount at L/S = 10 in the pH dependence test at the appropriate pH (around 7). The agreement between the different levels of testing (lab-lysimeter-field scale) point at solubility control as well. If solubility control can be demonstrated prediction of long term behaviour and identification of possible stresses at long term is quite well feasible.

Polycyclic aromatic hydrocarbons (PAH)

Besides the measurement of several major, minor and trace elements also organic contaminants were analysed in laboratory experiments, lysimeter and field leachate to verify to what extent approaches developed for inorganic constituents can be applied to organic contaminants as well. In Figure 9 the data for PAH from laboratory testing and leachate analysis in lysimeters and field are combined. PAH leaching increases as pH increases, which has been observed before (Comans, 2001). This is related to the association of PAH with DOC and particular the higher molecular weight humic type substances with a higher aromaticity. In the organic matter enriched integral waste mix the PAH leachability is higher.

In spite of the limited number of data presently available for PAH at the different levels, the observations are very promising indeed. The match

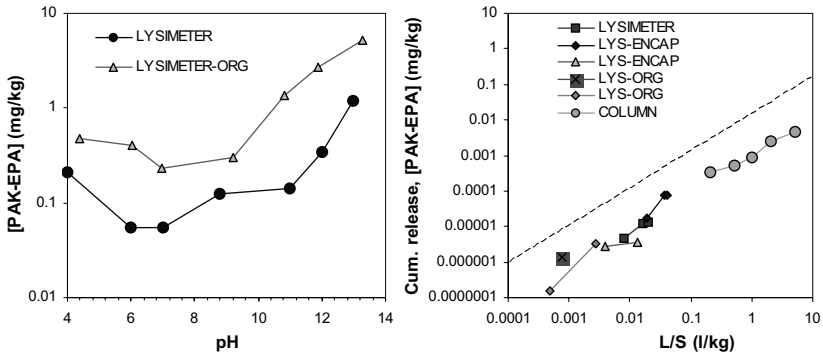


[Figure 8] Leaching of Pb as obtained from pH dependence test, percolation test, lysimeter and pilot cell leachate. Individual wastes are given in the plot on pH dependent leaching. The box represents most likely pH domain for the mixed waste. EU Landfill limits for non-hazardous waste inserted for comparison.

between the different levels of testing (lab-lysimeter-field) is encouraging. The observation that the cumulative release from a column test is significantly lower than a single step batch test at L/S = 10 resulting from more severe contact of waste and leachate, is confirmed here. This implies that a column test would be the more realistic procedure for judging organic contaminants. In the batch test, irreversible mobilization of DOC and colloids appears to take place leading to higher release than can be expected under slow percolation conditions. Although not further discussed here volatile organic compounds wash out like salts and show a consistent release level between batch and column as one would expect for non-interacting species. From other work (Roskam and Comans, 2003) it was found that release of PAH continues with L/S. However, the relevance of such releases must be balanced against the time scale for reaching such high L/S values. For a regular landfill scenario, L/S = 10 may correspond to 10,000 years of infiltration. Reducing the DOC level in leachate has direct consequences for release of organic contaminants.

Impact evaluation

A direct comparison of the landfill release data as obtained from characterization testing reveals that the predominantly inorganic waste landfill concept performs considerably better for many constituents than the criteria established for non-hazardous waste. In fact, for many constituents the release at L/S = 10 meets the inert waste criteria (Table 2). Cl and sulphate exceed the inert waste criteria. If preferential flow is considered (emission indicated with*), which may lead to a reduction in cumulative release at L/S = 10 of a factor of 3, then Cl is getting close to the limit. For sulphate, however, the limit is still exceeds the criterion. The reduction factor may not be applicable for sulphate as sulphate is solubility controlled. That implies that a fixed concentration will be measured for a long time until gypsum as a solubility controlling mineral is depleted.



[Figure 9] Leaching of PAH as obtained from pH dependence test, percolation test, lysimeter and pilot cell leachate. Dashed line represents slope 1 indicating solubility control.

The emissions of antimony are critical with respect to the acceptance of waste at an inert landfill site. The emissions in the pilot project are however continually slightly lower than the other experiments and the emission now seems to be levelling off. Subsequent measurements will indicate if this behaviour changes or if there is a temporary levelling off of the antimony emission. The antimony concentrations measured are generally lower than or around the level of the limit of detection (approximately 5 ppb with ICP-AES). More sensitive techniques than ICP-AES are available. These techniques were however not used in this research. Additional measurements are needed with a more sensitive technique (hydride generation and analysis with atomic fluorescence spectrometry) to establish if antimony is actually a critical element. On the basis of these results it cannot be unequivocally concluded that antimony is a critical element with respect to the acceptance criteria for inert waste. However, the reported antimony emissions can be seen as the upper limit for the antimony emission.

The chloride emissions exceed the standards for the acceptance of waste at an inert landfill site (even when corrected for preferential flow by assuming that also in the long term this part of the Cl will only come available very slowly and will then not pose a risk). A solution can only be found for this by reducing the total available chloride content, given that chloride is a mobile component that is not bound by the solid matrix. A quota system for this component can provide a solution so that the emissions comply with the current legislation for the acceptance of inert waste. The treatment of salt-containing wastes by cement-stabilisation and subsequent landfilling at a stabilised waste landfill site can also be considered. Another option is pre-treating chloride-containing waste streams with a sort of heap leaching process as an initial connected step for the removal of undesirable levels of mobile components. This could lead to considerably better control of the leachate quality for less manageable elements such as this. However, local groundwater conditions may need to be considered, as in locations close to the sea natural groundwater levels far exceed the criteria used for developing the Annex II criteria.

Sulphate appears to encounter problems in all landfill concepts. The solubility of gypsum gives rise to the problem; the solubility control ensures constant (too high) concentrations. This means that reducing the sulphate leaching by pre-treatment of wastes (washing or heap leaching) will have little effect on the quality of the wastes with respect to the leaching of sulphate. Besides, the solubility-controlling mineral even seems to control the leaching of sulphate to almost L/S = 10 (depletion has not yet occurred). The final fractions from the column tests do show that depletion can be expected around L/S values of 10 but these L/S values are much too high for pre-treatment procedures. An alternative for a number of sulphate-containing residues could be immobilisation using cement-stabilisation. However, the suitability of the waste stream concerned for this technique should be examined. Materials such as residues from construction and demolition waste segregation can be extremely heterogeneous and coarse, and thus for example unsuitable for immobilisation. The question is if this has consequences for the criteria, for the acceptance or that other solutions need to be found.

[Table 2] Comparison of measured release at L/S = 10 for the pilot Nauerna with inert waste criteria (EU LFD, 1999; 2003). (* Corrected for preferential flow).

Element/ parameter	Measured release in mg/kg at L/S = 10	EU LFD Inert in mg/kg at L/ S = 10	Element/ parameter	Measured release mg/kg at L/S = 10	EU LFD Inert mg/kg at L/ S = 10
pH	7.2	> 6	Hg	0.000007 (L/ S = 0.1)	
As	0.13	0.5	Mo	0.12	0.5
Ba	0.5	10	Ni	0.14	0.4
Cd	0.02	0.04	Pb	0.35	0.5
Cl	**3300 (1100)***	800	SO4	**11170 (3700)***	1000
Cr	0.019	0.5	Sb	**0.06**	0.06
Cu	0.038	2	Se	0.05	0.1
DOC	350	500	Zn	0.8	4

Conclusions

There is in general a very good agreement between the leaching behaviour of the constituted waste mixture and the leachate as obtained from the full-scale pilot experiment. This indicates that in spite of an apparently very heterogeneous mix of materials, leaching is governed by well-defined solubility controls for many constituents. The key aspect is to identify, which waste streams or external stresses can affect this balance in such a way that control is lost. Monitoring is continued to obtain a more extended overlap between testing at different levels.

Key controlling factors for this predominantly inorganic waste disposal are controls on the individual waste DOC levels (as shown with geochemical modelling), mobile inorganic (e.g. Cl, sulphate) and water-soluble organic contaminant levels. The behaviour of organic contaminants has been evaluated and shows a very consistent behaviour as well, in a similar data representation as used for inorganic constituents. In view of excessive DOC and colloid mobilization, a percolation test would seem more appropriate to assess organic contaminant release from a landfill than a single step batch leaching test (L/S = 10).

The main differences in emissions between the laboratory, lysimeters and the pilot experiment are caused by the different redox potentials measured (ranging roughly from -300 mV to + 300 mV) at the different scales of testing. The different redox potentials lead to relatively large deviations in the emissions of Cu, Fe and Mn on the different test scales.

Geochemical release modelling on column leaching test data showed that the model generally predicts the release of Cu, K, Pb and SO_4^{2-} very well. This implies that the chemical processes leading to release are understood. The results indicate that preferential flow aspects are important for the long-term prediction of the release of mobile elements from a landfill. The model predicts the release quite well. This modeling approach is comprehensive in that it deals with all major, minor and trace elements simultaneously and partitioned between dissolved (free and DOC bound) and solid phases (minerals, Fe-oxide, Al-oxide and particulate organic mat-

ter). However, a few contaminants show deviations in concentration of over one order of magnitude and these deviations need to be sorted out in future work.

The comparison of the cumulative release of mobile species in lysimeter and field situations with the laboratory characterisation of the mixed waste entering the landfill, gives an indication of preferential flow. Based on the present data, this may imply that only about 30 % of the cell is engaged in the leaching process.

A comparison of release as observed on the integral waste mix of pilot Nauerna shows that for all critical parameters the inert EU landfill criteria are met. An exception is formed by Cl and sulphate, while Sb is just on the limit. This observation indicates that already an important goal has been achieved. As inert waste does not require any form of lining, the conditions for minimum aftercare are approached.

Additional Sb measurements are needed with a more sensitive technique to establish if Sb is actually a critical element. On the basis of these results it cannot be unequivocally concluded that Sb is a critical element with respect to the acceptance criteria for inert waste. However, the reported antimony emissions can be seen as the upper limit for the antimony emission.

The chloride emissions exceed the standards for the acceptance of waste at an inert landfill site. A solution can be found for this by reducing the total available chloride content, which may not be easy to achieve. The treatment of salt-containing wastes by cement-stabilisation and subsequent landfilling at a stabilised waste landfill site can also be considered. In specific cases the local conditions need to be considered, as natural Cl^- levels may far exceed the target values used for setting the criteria.

Sulphate appears to encounter problems in all landfill concepts. The solubility of gypsum gives rise to the problem; the solubility control ensures constant (too high) concentrations. This means that reducing the sulphate leaching by pre-treatment of wastes (washing or heap leaching) will have little effect on the quality of the wastes with respect to the leaching of sulphate. The final fractions from the column tests do show that depletion can be expected around L/S values of 10 but these L/S values are much too high for pre-treatment procedures. An alternative for a number of sulphate-containing residues could be immobilisation using cement-stabilisation. The question is if this has consequences for the criteria, for the acceptance or that other solutions need to be found.

At the Nauerna location it is possible that the high chloride and sulphate emissions do not present a problem in practice because the landfill site is located in an area with high salt concentrations in the groundwater. A decision on the acceptable long-term emission levels may be actualized by a comparison of the predicted impact and actual concentrations in the groundwater. This situation must be discussed with the competent authority so that further clarification is obtained. Annex II of the European Landfill Directive does not give room for a location-specific risk assessment.

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