

**PREDICTION OF LONG TERM LEACHATE QUALITY
AND CHEMICAL SPECIATION FOR A
PREDOMINANTLY INORGANIC WASTE LANDFILL**

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PREDICTION OF LONG TERM LEACHATE QUALITY AND CHEMICAL SPECIATION FOR A PREDOMINANTLY INORGANIC WASTE LANDFILL

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SUMMARY: The systematic approach to evaluate the sustainable landfill concept for predominantly inorganic waste by comparing release at different scales of testing (lab-lysimeter and field) gives a clear insight in the release controlling factors and the long term leaching behaviour. The relationships found and the conclusions that can be drawn for important inorganic and organic parameters imply an important step forward. The realization that a waste mix behaves more consistently than one would expect based on the obvious heterogeneity indicates is striking and holds promise for future control of leachate quality. For the sustainable landfill concept of predominantly inorganic waste, it is already clear that for a large number of parameters, inert waste criteria as specified in the EU Landfill Directive can be met.

1. INTRODUCTION

The study of the Sustainable landfill concept for predominantly inorganic waste has been addressed at previous conferences (van der Sloot et al, 2001; Van der Sloot et al, 1999a). The study integrates testing at laboratory, lysimeter and pilot scale with modelling of long term release. Emphasis is placed on understanding the processes within the landfill as the mutual interaction of individual wastes determines the ultimate long-term quality of the leachate (Van der Sloot et al, 1999b). The anticipated outcome is a disposal practice through waste acceptance that will reduce the contaminant emissions to acceptable levels on the long-term; thus reducing long-term aftercare.

In the mean time more leachate data from the lysimeters at the test site in Petten and from the pilot at landfill Nauerna have been obtained, which allow a more elaborate comparison than before. In addition, organic contaminants have been measured in laboratory leaching experiments as well as in leachate from the lysimeters and in leachate from the 12,000 m³ pilot.

The key question remains: is leachate treatment required in the long-term? In this paper the systematic leaching behaviour of a waste mix and release controlling mineral phases will be addressed. The relationship between different levels of testing (lab-lysimeter-field) is evaluated after 3 years of data collection. The performance of the predominantly inorganic

waste concept for sustainable landfill is evaluated against current criteria and targets of minimum aftercare.

2. MATERIALS AND METHODS

2.1 Laboratory experiments, lysimeter and field measurements

In the framework of a Dutch national research project on sustainable landfill, laboratory experiments (percolation test – PrEN 14405, 2003; pH dependence test – PrEN 14429, 2003), lysimeter studies (1-1,5 m³) and a 12,000 m³ pilot demonstration project at landfill site Nauernasche Polder (the Netherlands) have been 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 PrEN 14405 and PrEN 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; Lysimeter 2: Disposal of wastes in order of delivery as practiced at Nauerna with addition of 5 w/w % of sewage sludge and car shredder waste each to increase organic matter loading (Coded LYS-ORG); Lysimeter 3: Encapsulation of more contaminated wastes in relatively low permeability wastes (soil cleaning residues; also coded LYS-ENCAP).

The studies at field, lysimeter and laboratory scale represent different time scales through the liquid to solid ratio, to which the waste was exposed.

2.2 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, TI, V, Zn). DOC and TIC were analysed by a Shimadzu TOC 5000a analyser. Cl, F, ammonium and sulphate were analysed by ion-chromatography. PAH was analysed by HPLC, mineral oil and volatile organic halogenated compounds were analysed by GC-MS.

2.2 Geochemical speciation and release modelling

The role of minerals and dissolved organic carbon (DOC) on metal mobilisation is addressed by applying the NICA-Donnan model (Kinniburgh et al., 1999) in a new modelling environment ORCHESTRA (Meeussen, 2003) for geochemical solubility calculations using an extended MINTEQA2 database. For this purpose ORCHESTRA is coupled to a

database/expert system (van der Sloot et al, 2003a; van der Sloot et al, 2003b) 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 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; 2003) allows a verification of performance of the new landfill concept against criteria.

3. RESULTS AND DISCUSSION

3.1 Leachate production

The natural precipitation is monitored at the lysimeter site. In figure 1 the cumulative precipitation and the cumulative leachate production per lysimeter are shown as a function of time. The precipitation varies per lysimeter as the surface area is different for each, which obviously affects the leachate production per lysimeter. For lysimeters 2 and 3 the difference in cumulative precipitation is small. However, the difference in cumulative leachate production is significant, which is probably due to the incorporation of dried sewage sludge (94 % d.w.) and relatively dry shredder waste (95 % dw). These wastes take up water before net production of leachate will occur. During the period October to March leachate production is about half of the net precipitation. However, during April to September evaporation balances net precipitation. After an initial delay in leachate production, lysimeter 2 has caught up with lysimeter 3 in terms of leachate production. For long-term prediction purposes, this type of information is important.

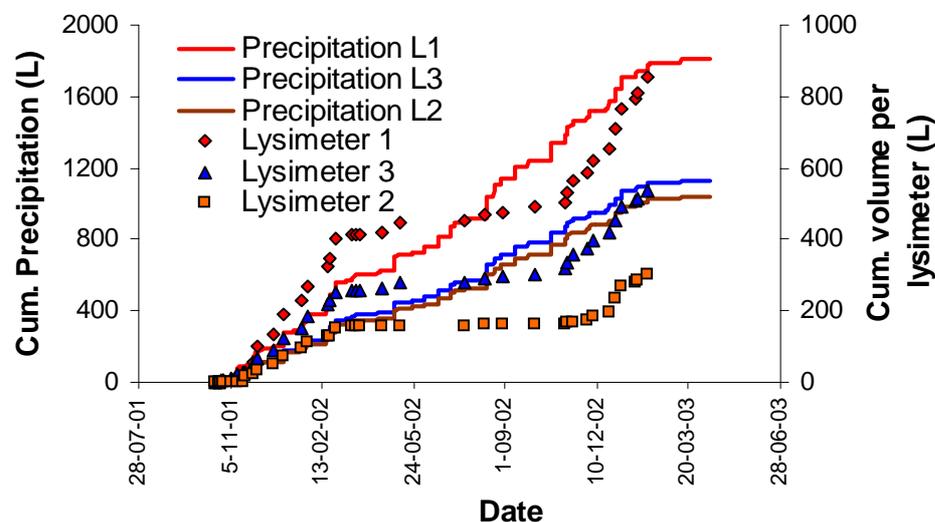


Figure 1: Cumulative precipitation and cumulative leachate production per lysimeter as function of time.

3.2 Dissolved Organic Carbon

In Figure 2 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 (van Zomeren et al, in preparation). 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 sub-samples 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.

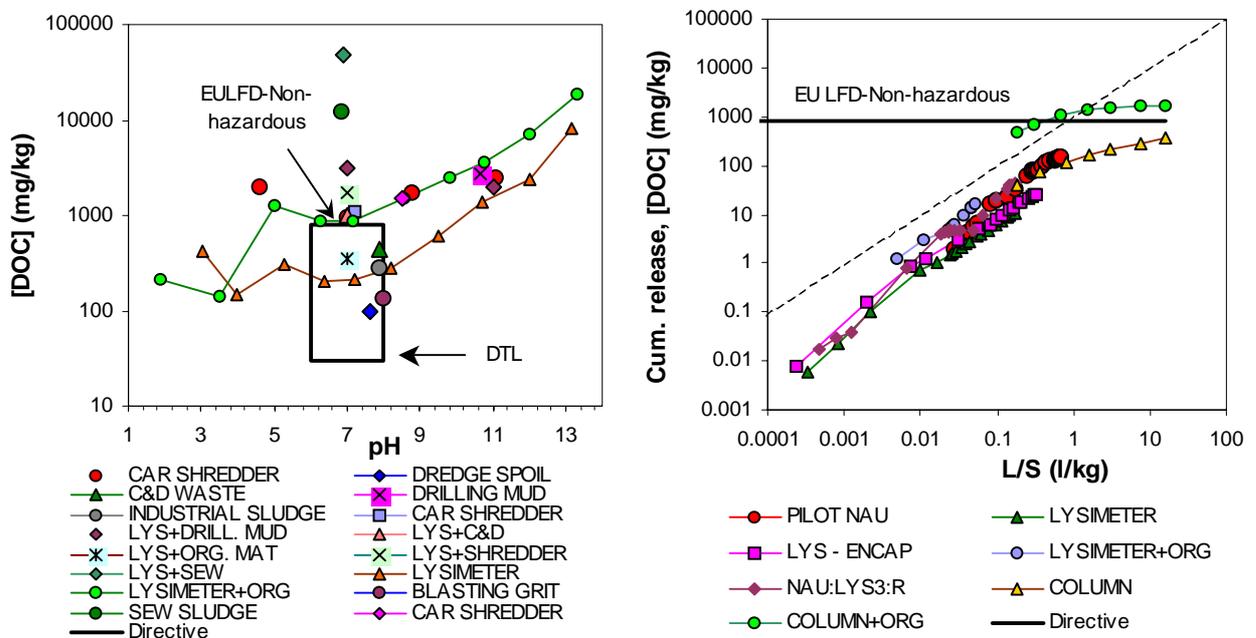


Figure 2: Leaching of DOC as obtained from pH dependence test, percolation test, lysimeter and pilot cell leachate. Individual wastes are given in the plot on pH dependent leaching. Box represents most likely pH domain for the mixed waste. DTL = detection limit. EU Landfill limits for non-hazardous waste inserted for comparison.

3.3 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 up-flow (minimal channelling) will allow conclusions to be drawn on the possible role of preferential flow. In Figure 3 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.

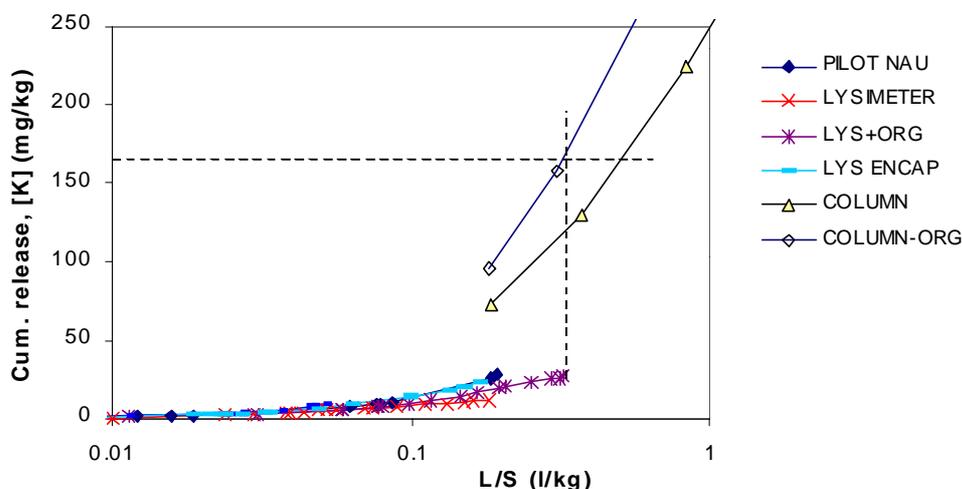


Figure 3: Comparison of release of K derived from lysimeter and field leachate data with results from up-flow column leaching tests.

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 3 and Table 1 indicate that about 30 % of the mobile species is released at a given L/S from lysimeter and full-scale systems relative to the release obtained for column leaching tests. This implies that more than 60 % of the waste is not affected by infiltrating rainwater.

3.4 Lead

In Figure 4 the data for Pb from laboratory testing and leachate analysis in lysimeters and field are combined. The leaching behaviour of Pb 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. If solubility control can be demonstrated, prediction of long term behaviour and identification of possible stresses at long term is quite well feasible.

3.5 Polyaromatic hydrocarbons

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 5 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 et al, 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.

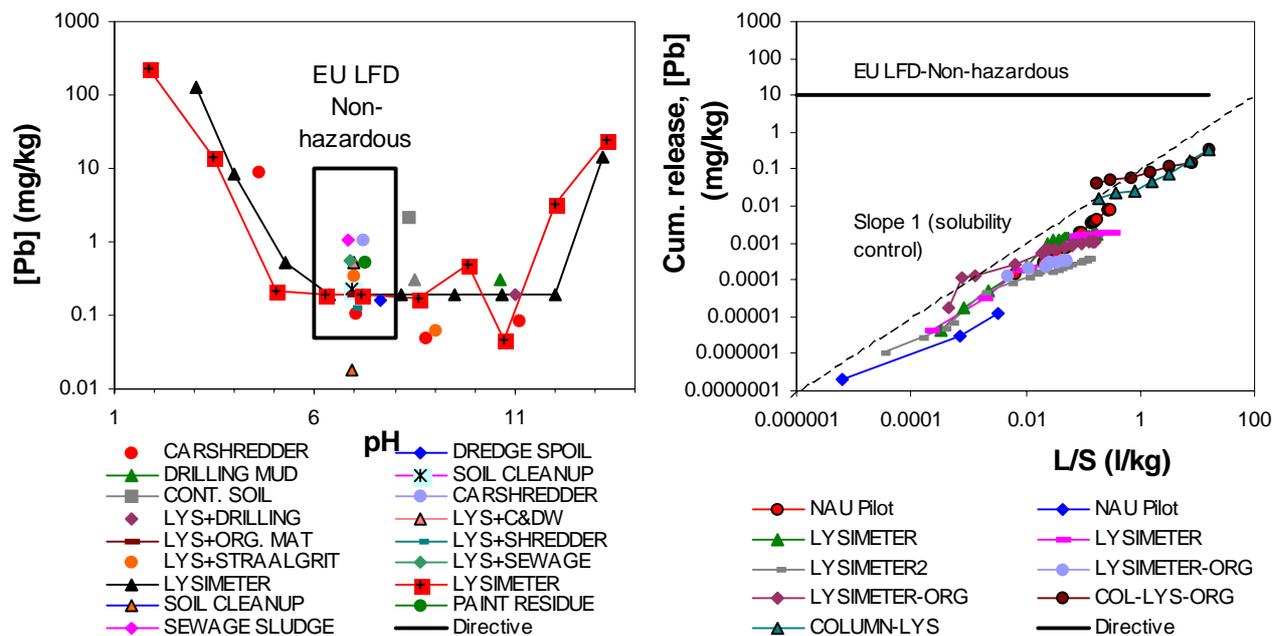


Figure 4: Leaching of Pb as obtained from pH dependence test, percolation test, lysimeter and pilot cell leachate. Individual wastes are given in the plot on pH dependent leaching. Box represents most likely pH domain for the mixed waste. EU Landfill limits for non-hazardous waste inserted for comparison.

In spite of the limited number of data presently available for PAH at the different levels, the observations are very promising indeed. The match 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 $L/S=10$ (Grathwohl et al, 2001) 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.

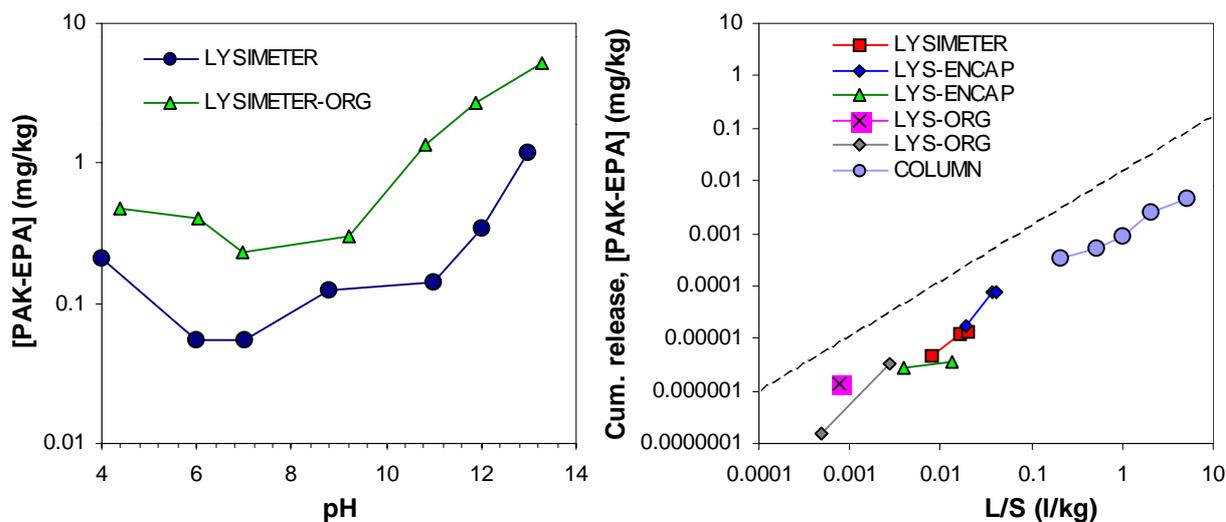


Figure 5: Leaching of PAH as obtained from pH dependence test, percolation test, lysimeter and pilot cell leachate. Dashed line represents slope 1 indicating solubility control.

3.6 Geochemical modelling and release modelling

Using the new modelling environment ORCHESTRA (Meeussen, 2003) for geochemical solubility calculations with an extended MINTEQA2 database coupled to an Access database/expert system, the results of laboratory test data, lysimeter data and field leachate data can be evaluated simultaneously. Although the speciation has been calculated for many major, minor and trace elements, there is only room for a few examples here.

In Figure 6, the modelling results for Pb and Mo are given. It is striking to note that the mineral PbMoO_4 appears to be solubility controlling over a wide pH range (pH 2-8) and a very wide L/S range (0.0001 to 10). This is rather extraordinary and points at a great relevance of this particular mineral phase in this complex mixture, containing a wide range of waste materials. At pH above 10, the mineral $\text{Pb}(\text{OH})_2$ is relevant as it fits the pH dependence test data well. Above pH 10 no other relevant phases are identified as the solution is undersaturated with respect to CaMoO_4 . For Pb and Mo, complexation with DOC was not a main release mechanism resulting from the NICA-Donnan model incorporated in the speciation calculations.

The capability of calculating the chemical speciation of eluate data generated by the pH dependence test (pH variation), a percolation test (L/S variation) and leachate data (lysimeter and field) simultaneously, offers the possibility to draw conclusions on a large number of practical situations and time scales.

Release modelling using relatively simple models is feasible for several constituents. For elements involved in substantial chemical changes in solubility control more sophisticated modelling is needed (Dijkstra et al, 2003).

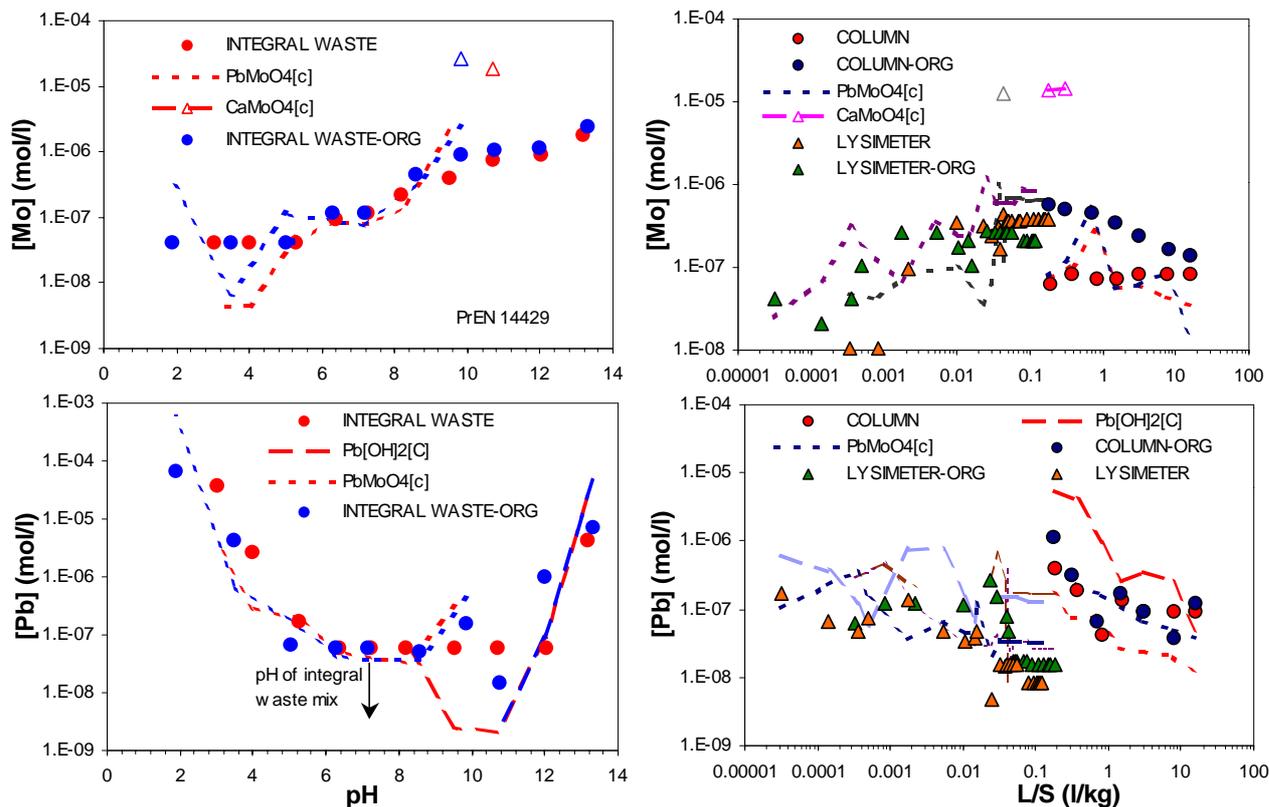


Figure 6: Geochemical modelling results for lead and molybdate for the different waste mixes tested in laboratory experiments and lysimeters.

3.7 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, whereas Sb is at the limit. If preferential flow is considered, 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 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.

4. CONCLUSIONS

The mutual consistency of the data at different scales (not only for DOC, Pb, Cl and PAH, but for several other parameters also) as visualized by a specific data representation, has been reconfirmed with more data and forms a very promising approach to tackle the issue of assessing long-term leachate quality and the means to control it by a new waste acceptance policy. Monitoring is continued to obtain a more extended overlap between testing at different levels.

It is striking to note the agreement between the leaching behaviour of the constituted waste mix and the leachate as obtained from the full-scale demonstration. This indicates that in spite

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; ** At L/S=0.1).

| 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 in mg/kg at L/S=10 |
|-----------------------|---|--|-----------------------|-------------------------------------|--|
| pH | 7.2 | > 6 | Hg | 0.000007 ** | |
| 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 | SO ₄ | 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 |

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.

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, sulfate) 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 geochemical speciation modelling allows for a direct comparison of solubility control at a wide range of pH and a wide range in L/S as well as in field leachate, providing a means of gaining more understanding and subsequently more control over long-term release.

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.

The ammonia release from the lysimeters is significantly less than the data from field leachate and lab test do indicate. The role of nitrifying bacteria in the lysimeter set-up must be verified. The aerobic conditions in the collection device may enhance conversion of ammonia to nitrate.

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.

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