Opening the Black Box

Process-Based Design Criteria to Eliminate Aftercare of Landfills

Dutch Sustainable Landfill Foundation

Colophon

STICHTING DUURZAAM STORTEN

Content

1.   Introduction

1.1   Background

Landfilling will always be needed in order to dispose of wastes which cannot be recycled or treated in other ways. The processes occurring within landfill bodies are determined by the large heterogeneity of disposed materials. At the start of this project the understanding of these processes was insufficient to enable reliable predictions of releases to air, soil and groundwater to be made. In 1998, the Dutch Association of Waste Management Companies initiated the establishment of the Dutch Sustainable Landfill Foundation. The Foundation aims to develop sustainable landfill technologies based on an understanding of the chemical processes occurring in landfills. The work presented here covers the results of the Dutch Sustainable Landfill Project.

Currently, legislators consider landfill bodies to be black boxes from which all manner of unwanted emissions occur. This 'black box thinking' has resulted in the adoption of precautionary measures aimed at protecting the environment. These require the material in landfill bodies to be eventually contained in an impermeable shell so that emissions are prevented. During the period that the landfill is in use, leachate (percolate) and gases produced within the landfill are collected and cleaned or utilised. From the containment perspective, wastes are assessed on a waste-by-waste basis – that is each type of waste from each client is assessed individually. There is no evaluation of the integral waste mix which constitutes the landfill body. This is a serious limitation when designing new landfill concepts that focus on low releases in the long-term.

The European Union's Landfill Directive (Landfill Directive) (*1*) recognises three different types of landfill: for inert, non-hazardous, and hazardous waste. Specific measures need to be taken for each type of landfill. Annex II of the Landfill Directive (*2*) is a first step towards 'a source term definition' for controlling emissions to groundwater; that is the total emissions through the landfill liner into the groundwater at any given time are considered. However the current Landfill Directive has limitations. The landfilling of hazardous waste in accordance with the acceptance criteria specified in Annex II requires eternal aftercare, as leachate quality is not likely to meet the quality objectives of the Water Framework Directive. The WFD (Water Framework Directive) puts requirements on all activities affecting soil and groundwater (primarily groundwater). Therefore, future emission levels from landfills will be derived from the WFD.

Acceptance assessments on individual wastes, without taking the overall effect of the waste mix into consideration, is not an approach that provides the answers needed for the evaluation of long-term emissions from landfills. For example, desirable waste–waste interactions are prevented and undesired waste-waste interactions can occur (such as the formation of hydrogen gas caused by the disposal of fine metallic aluminium which comes in to contact with a highly alkaline waste). The fact that these undesirable waste interactions can occur is because each of these wastes may individually pass the legislative acceptance criteria but the combination causes problems. Such issues need to be discussed by the Technical Adaptation Committee (TAC) during the revision of the Landfill Directive.

Landfill containment seems an environmentally safe option but it requires a large investment and a long-term commitment; especially for the long-term maintenance of the landfill cap. The precautionary measures beneath a landfill are impossible to maintain after the operational life of the landfill. Therefore, there is no guarantee of the performance of a bottom liner for eternity. The potential for harmful emissions will remain as a result of containment and they may be released at some unknown point in the future, when control measures are no longer operational.

1.2   Aims of the project

This report presents the main features and results of an investigation into possible alternative approaches to reduce and prevent long-term emissions from certain types of landfill. The overall aim of this project has been to generate the knowledge and the technology needed to construct landfills, which after an operational period with active control (preferably less than one generation) can be considered inherently safe because releases to the environment are down to a level such that long-term aftercare is not needed. This implies that the concentration levels of the constituents released are below critical levels, which at this point in time are defined as the condition that corresponds to the criteria specified for the landfilling of inert waste. The justification for this is that the EU does not require any form of additional protection or control for this type of landfill and thus the emissions can be considered to present an acceptable risk to society.

The fundamental idea behind this project is that knowledge of the processes responsible for the harmful emissions will lead to technological and design measures being developed that will allow the control of processes that cause emissions. In addition better use of those naturally occurring processes that reduce emissions could also be made.

This report intends to show that, within the timeframe of one generation:
- Concentrations of all relevant pollutants in leachate can be reduced to levels below legislative standards.
- Concentrations are not likely to increase again after this time.
- The emissions that do occur cause no adverse effects on soil and groundwater.

In the project the behaviour of the organic macro chemical parameters (biological oxygen demand (BOD), chemical oxygen demand (COD), organic nitrogen (N_{Kj})), the organic micro chemical pollutants, major and minor elements (e.g. Ca, Al, Si, Mg), metals, oxyanions (e.g. Mo, As, Sb, V) and salts were investigated. A number of processes were studied, in order to find the means of controlling emissions. These were: waste selection, enhanced biodegradation, immobilisation, removal (flushing), and solubility control. Instead of investigating each process separately, all the aforementioned processes were integrated into a consistent framework.

The approach adopted in the project was to demonstrate the occurrence of processes that have a major impact on emissions (including those that can be optimised to reduce emissions), in experiments carried out on different scales (laboratory, lysimeter and field). The results from these experiments are interpreted in a generic framework based on the modelling of the fundamental hydrological, biological and geochemical processes occurring in landfills.

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1.3   Project

This project was initiated and funded by the Dutch Sustainable Landfill Foundation (NV Afvalzorg , A&G Maasvlakte, Essent Milieu, Stainkoeln and Vereniging Afvalbedrijven (Dutch Association of Waste Management Companies)).

2.   Theory

2.1   Emissions from landfills

The landfilling of wastes gives rise to various emissions. The main processes responsible for the release of contaminants are leaching with infiltrating water and emissions of gases, resulting from the biological degradation of organic matter to CH_4, CO_2 and water.

Organic macro components are generally expressed as BOD, COD and organic-N (N_{Kj}). They consist of macromolecules such as carbohydrates, proteins and fats, both solid and dissolved, which are converted into smaller components as sugars, amino acids, fatty acids. These smaller components are ultimately converted into acetic acid, N_{Kj} and biogas. During this biological process part of the carbohydrates, proteins and fats are used for growth by the micro-organisms, resulting in fixation of these components in biomass. The biochemical processes that result in the formation and decay of organic macro components are reasonably well established, and the overall process is often described in four, five and sometimes even nine phases: hydrolysis, acetogenesis, various phases of methanogenesis, and sometimes (partial) mineralisation under aerobic conditions (*3*).

However, the cascade of reactions is complicated by the heterogeneity of the waste and in practice different phases exist simultaneously throughout the landfill. The rate of decay depends on a number of factors, e.g. waste composition, moisture content and temperature.

Organic micro components are generally already in the waste upon deposition, and may end up both in the gas and the leachate. Organic micro components can be converted or decomposed under both anaerobic and aerobic conditions, sometimes generating other even more troublesome pollutants, e.g. vinyl chloride from hydro-dechlorination of tri-chloroethene.

Metal ions are also present in the waste upon deposition. However, the majority of metals stay immobilised and do not end up in the leachate. Mobilisation and immobilisation of metal ions occur in a number of processes, which are dependent on pH and the extent of degradation of the organic matrix, as indicated in Figure 1. The fully drawn top line (in brown) represents the total amount of the component under consideration present in a particular waste mix; not all of this is available for leaching. The blue dotted vertical line, marked 'potentially leachable', represents the maximum amount that can be leached. The curved red line indicates the amount that is actually leached from the pure material, dependent on the pH. The actual leaching in a landfill environment can be different from the behaviour of the pure material, as a result of a number of processes. The actual leached amount can be lower than expected as a result of sorption onto organic materials and the occurrence of reducing conditions. On the other hand, leaching can be increased as a result of chloride and dissolved organic carbon (DOC) complexation.

The pink frame around neutral pH, indicates the most likely circumstances that can be reached by manipulation of the landfill design and operation, and specifically through the presence of a sufficient amount of stable organic material.
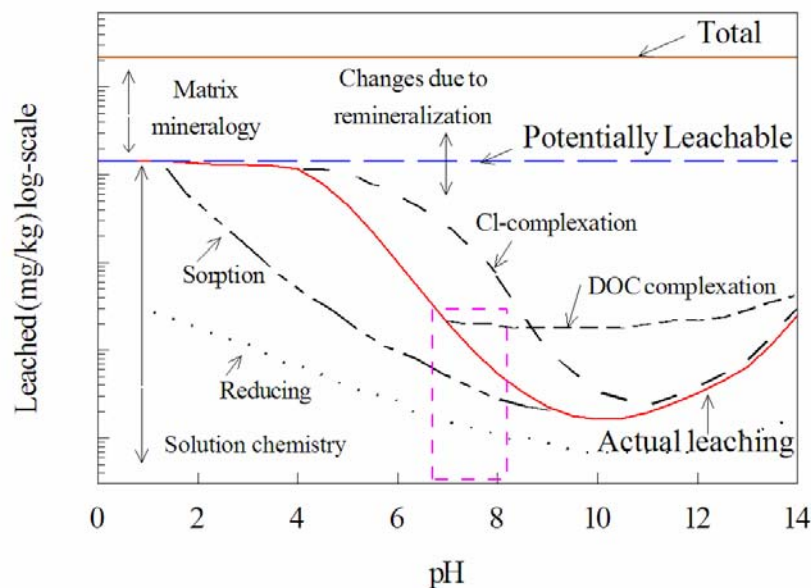
Figure 1   Overview of processes occurring in landfills that have a major influence on the leachability of contaminants to the surroundings

Oxyanions such as SO_4^{2-}, CO_3^{2-} and PO_4^{3-} come from sulphur and phosphorus-precursors, present in the waste upon deposition. The amounts of oxyanions available for leaching are largely determined by the waste composition. The same goes for **salts** such as Cl^-.

2.2   Reducing emissions from landfills

Emissions from landfills to the atmosphere (e.g. CH_4, CO_2) and releases of both organic and inorganic contaminants from landfills to soil, surface water and groundwater can be reduced in a number of ways:

- **Waste selection**: This can be used in three different ways:
   * Prevention: not allowing the contaminant to enter the landfill in the first place.
   * Adding/preventing components to the waste to enable certain processes to occur in the waste, e.g. using permeable shredded tyres rather than more impermeable daily covers to enable flushing.
   * By combining materials in such a way that through their interaction the emission potential for all contaminants is reduced. The interaction may be directly between waste components but the combination of materials may also create the right conditions in which other reactions can be stimulated, e.g.:
      o Adding buffering components to enhance methanogenesis.
      o Stabilisation of solid organic material results in a reduction of dissolved metal ions due to complexation to the solid matrix.

- **Pre-treatment**: Pre-treatment can aim to remove specific components prior to landfilling. Examples are the mechanical removal of metals, paper or plastics; washing to remove salts or biological pre-treatment (aerobic or anaerobic biodegradation) to convert part of the biodegradable organic material. Certain pre-treatment processes might also be used to enable or promote subsequent processes, e.g. shredding to enhance biodegradation or to facilitate flushing.

- **Biodegradation/Conversion**: This is similar to pre-treatment and can also occur in the landfill itself. Organic macro components are formed but also largely converted during biodegradation. Conversion can be enhanced through leachate infiltration or by creating an aerobic environment in which composting processes occur rather than fermentation. Many organic micro components are also converted under anaerobic conditions. It is likely that measures that enhance the biodegradation of organic material, also enhance the degradation of organic micro components. Biodegradation also affects heavy metal concentrations in the leachate, as dissolved organic carbon concentrations are reduced, thereby mobilising metal ions by complexation.

- **Immobilisation**: This can be used as a pre-treatment process to facilitate the landfilling of materials that would otherwise not be suitable for landfilling, by binding contaminants in mineral or other phases in the landfill. Matrix retention occurs as less soluble minerals develop in the landfill body by sorption onto solid organic matter (residual fraction), mineral oxides and other charged surfaces such as are present in clay minerals.

- **Solubility control**: The release of metal ions is determined by the distribution between the solid and the liquid phase. The partitioning in turn is mainly determined by the pH, conductivity, redox and DOC concentrations in the liquid phase (see Figure 1). A minimum level in the liquid concentrations can be achieved by controlling these circumstances and processes. If the release of a certain component is determined by solubility control, the application of flushing is only capable of reducing its concentration if the flushing is continued. When the flushing stops, the system will return to its equilibrium concentrations; at least for as long as there is enough material left in the solid phase.
  Conversion of the component to the right chemical form can be achieved by combining different waste batches in an optimal way, or by adding specific chemical or mineral agents to allow the component to precipitate in the most suitable form. The addition of specific chemical/mineral agents was not undertaken as part of this study.

- **Flushing**: Of the processes discussed flushing is probably the most straightforward to understand. The principle behind flushing is that soluble components are removed from the liquid phase of the landfill by introducing fresh liquid that does not contain the component in question. From a theoretical point of view it is clear that a reduction of the concentration by two orders of magnitude will require a throughput of fresh liquid in the order of three times the liquid content of the landfill. If this is to be achieved by natural precipitation, the timescale of one generation is not sufficient. In order to reach sufficient dilution in the proposed time period the infiltration rate has to be in the order of 1,500 – 3,000 mm/yr. This puts certain demands on the construction and operation of the landfill:
  * The permeability of the landfill body has to be high enough to allow a water flow of this magnitude. The permeability of the landfill body is a function of several parameters including waste composition, form of compaction, homogeneity, height of the landfill and the build-up of the landfill in layers.
  * In order to avoid having to discharge large amounts of leachate and infiltrate comparable amounts of clean water at the same time, a recirculation loop, which includes specific treatment of the leachate, is necessary.

2.3   Towards an integrated research programme

The composition of the waste that is supplied to a landfill largely determines the principal landfill processes and the landfill's pollution potential. This study considered three sustainable landfill concepts, each displaying a specific kind of reactive behaviour, which require different types of measures to control or manipulate the bio- and geochemical processes occurring within the landfill body. The three types are:

- Landfills containing predominantly organic waste. In these landfills biodegradation is the principal mechanism determining pollution potential.
- Landfills containing predominantly inorganic waste. In these landfills solubility control and leaching are the predominant factors determining the pollution potential.
- Landfills containing hazardous waste, where immobilisation can be an important mechanism to retain hazardous pollutants in the waste matrix.

The hypothesis in this study (see Figure 2) is that (i) by controlling processes, emissions from both the predominantly organic waste landfills and the hazardous waste landfills can be transformed into those of a landfill with the characteristics of an inorganic waste landfill and (ii) through solubility control and flushing, emissions from this inorganic waste landfill can be reduced further to those of a sustainable landfill with negligible emissions.
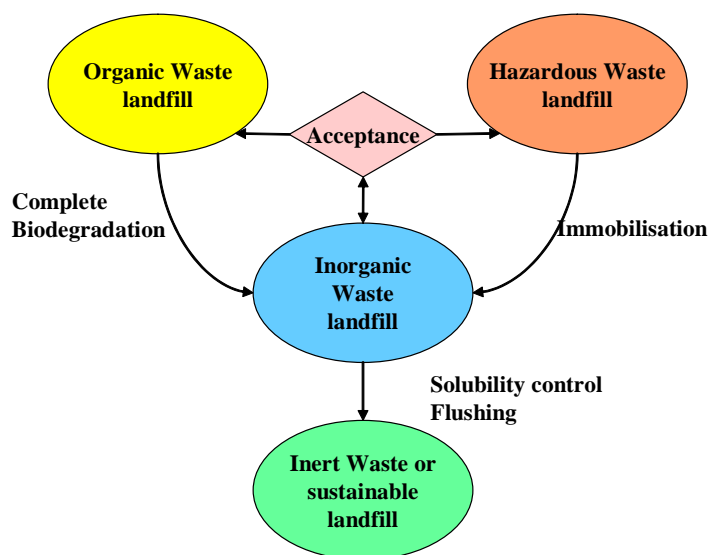
Figure 2   From landfills containing predominantly organic waste or hazardous waste to a landfill resembling one containing predominantly inert waste and beyond towards a sustainable landfill

Figure 3 describes a typical metal ion concentration in the leachate during the transformation process from an organic waste landfill to an inert waste landfill and subsequently to a sustainable landfill. In the organic waste landfill, biodegradation results in high concentrations of dissolved organic material in the leachate. Metal ion concentrations are also high in the leachate, as complexation to dissolved organic materials is the determining mechanism for metal ion dissolution. The amount of dissolved organic material is reduced as biodegradation progresses, until complexation is no longer the most important factor determining metal ion concentrations. At this point, the landfill has the properties of an inorganic waste landfill, where

emissions are governed by solubility and flushing. Further flushing ultimately results in concentrations that can be considered sustainable.

Figure 3   Development of leachate concentrations as a function of time for a conventional organic waste landfill, a sustainable organic waste landfill and a sustainable inorganic waste landfill. A sustainable organic waste landfill will tend towards the behaviour of a sustainable inorganic waste landfill.
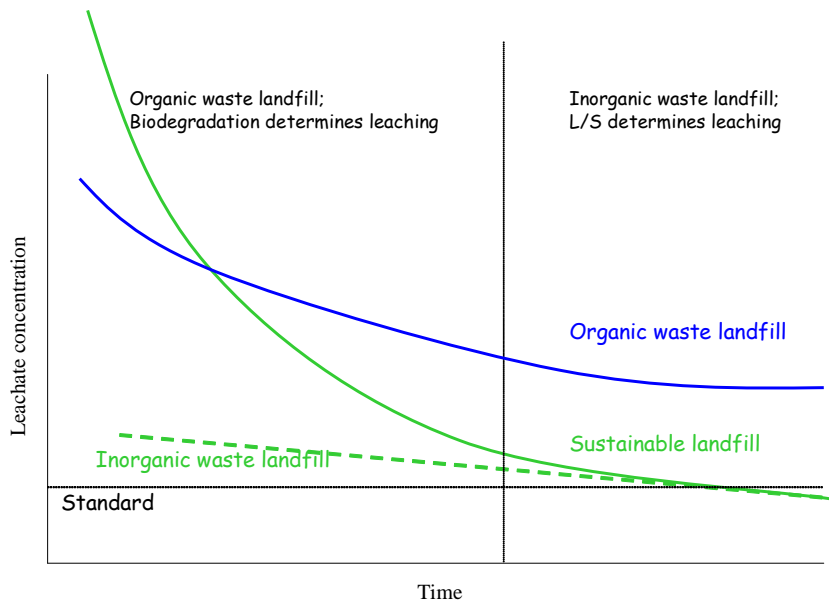
2.4   Landfills containing predominantly organic waste

The emission behaviour of waste containing high amounts of degradable organic matter is dominated by the degradation of organic matter. Landfill gas is produced during biodegradation and concentrations of BOD, COD and N_{Kj} in leachate are high in this characteristic phase of this type of landfill. The high dissolved organic matter content in the leachate also results in high amounts of metals in the leachate. When biodegradation reaches completion, leachate concentrations are significantly reduced. Over time the degradable organic matter in the waste is stabilised to non-degradable organic matter and the behaviour of the landfill has evolved into that of a landfill containing predominantly inorganic waste. The target is to ensure full degradation of the degradable organic waste fraction in this type of landfill so that the more stable end condition of predominantly inorganic waste landfills can be reached within one generation. Leachate recirculation or the creation of conditions for aerobic conversion are the primary control measures needed to achieve complete degradation at full scale.

2.5   Landfills containing predominantly inorganic waste

For this type of waste, pH can have a large effect on leaching behaviour. In a predominantly inorganic waste landfill the release of substances is determined by the partitioning of contaminants between the solid phase and leachate. Understanding the factors controlling this partitioning during the lifetime of a landfill holds the key to controlling leachate quality. pH, conductivity, redox and DOC are determined by the major elements and nature of the organic matter in the waste. Therefore, the latter are important factors in determining the degree to which contaminants are partitioned between the solid and liquid phases and also between free

and complexed forms. The interaction of both inorganic and organic contaminants with DOC is crucial in this respect, as the complexed forms are more mobile and in some cases less accessible to micro-organisms.

Although wastes are assessed on a waste-by-waste basis upon delivery to the landfill, in line with Annex II of the Landfill Directive, it is of great importance to know how releases from a landfill body can be described. The interactions of wastes in a landfill body have the tendency to develop a mixed waste release behaviour that can be quantified and described (*4*).

Describing behaviour, and thus understanding the controlling factors, provide management options to deal with undesired release behaviour. Currently, modelling capabilities have developed to such a level that meaningful conclusions on the behaviour of mixed waste can be drawn.

2.6   Landfills containing Hazardous waste

The leaching behaviour of hazardous waste should be reduced as far as possible. Therefore, the emphasis should lie in modifying the release behaviour of the widely varying waste qualities in this category by creating a chemically and physically more homogeneous, and thus better controlled, matrix. A major challenge is to ensure that such target conditions are not far from equilibrium with the landfill's surroundings and can contribute to the sustainable character of the landfill. This is ensured by a landfill design that enhances natural sealing and buffering of leachate pH by the inclusion of a soil layer.

There is a wide body of literature supporting the relevance of the above described processes. Generally speaking neutral pH levels, slightly reducing conditions combined with low DOC levels (especially in the form of humic and fulvic acids) tend to lead to relatively low emissions of metals and organic contaminants from any of the aforementioned landfill concepts.

Stabilisation of hazardous waste can be undertaken in various ways. Recipe development and the control of a stable and sustainable end product are main objectives. Work by Ludwig *et al*. (*5*) and by Fitch and Cheeseman (*6*) on cement-stabilised municipal solid waste incinerator (MSWI) fly ash (as large monolithic waste blocks) has shown surface deterioration. In both studies carbonation (precipitation of $CaCO_3$) was noted. In the studies by Baur *et al*. (*7*) and Fitch and Cheeseman (*6*) a very high pH in the leachate was observed, as would be expected for the highly alkaline matrix.

Carbonation is believed to be an important process that results in the sealing of pores. This implies that there is a lower release of contaminants due to the reduced diffusion speed. Once there is a proper understanding of the major processes and their significance in the release of contaminants from such waste materials, the behaviour of a monofill can be predicted by geochemical/transport models.

3.   Methods

3.1   Experiments on different scales

The approach taken in studying the development of new landfill concepts was to combine (i) experiments on individual wastes and mixed wastes on the laboratory scale, (ii) on the lysimeter scale and (iii) pilot studies on the field scale. Experiments were carried out for the same waste mix on all scales. The results from all the experiments were interpreted in a general data interpretation and modelling framework which allows an integrated assessment of the relevance of certain processes on all scales to be made. It also enables extrapolation of the data to real scale landfill scenarios.

Basic characterisation undertaken by laboratories covers a wide range of exposure conditions and has formed the basis for modelling chemical speciation, degradation of organic matter and chemical reaction/transport (taking preferential flow into account). The integrated approach used in this study gives an insight into possible long-term changes in behaviour. When placed in context with actual landfill leachate data it allows conclusions to be drawn on the reduction in emissions that can be achieved. The rationale of testing on different scales is that each scale provides an essential piece of the puzzle, as illustrated in Figure 4. Full scale testing can never provide the information needed for long-term projections. On the other hand laboratory testing cannot provide an insight into the key factors controlling releases under full scale conditions.

The geochemical modelling approach was designed to be applicable to any material or matrix by incorporating all the possible chemical and physical release control factors: mineral precipitation/dissolution, sorption onto iron and Al oxides, interaction with dissolved and particulate organic matter, and physical aspects such as permeability, tortuosity, preferential flow, etc.

Insight into long-term emission behaviour and methods to influence long-term emissions are of the utmost importance, both for the detailed design and evaluation of sustainable landfills. Full scale demonstrations only give insight into the development of emissions in the first few years. Column experiments or leaching tests have the potential to provide more information on long-term emissions of reaction products.

However, the speed of some essential processes is determined by their intrinsic kinetics and cannot be speeded up in the laboratory. Besides, real life emissions are effected by the fact that conditions in reality are less than ideal and not homogeneous. Modelling is used as a tool to facilitate the evaluation of long-term behaviour, by extrapolating the results of field trials, column experiments and leaching tests to real life long-term emissions.

Leaching and other characterisation experiments were carried out on different scales with different mixtures of waste materials. The experiments carried out on the laboratory scale were primarily focused on characterising the different types of waste materials that constitute the landfill mix. The characterisation experiments were: batch equilibration tests, pH-stat experiments, and column experiments. The function of the pH-dependent leaching test is to

understand the chemistry and the effects of biochemistry on the system. The laboratory scale column test provided a long-term projection of release behaviour that could not be obtained from lysimeter or full scale pilot studies. However, the combination of testing at these levels allowed important differences between the experiments to be identified (e.g. measure for preferential flow). From the behaviour of many constituents it is obvious that the target pH for a landfill of mixed granular wastes should be around 7 to 8, as this condition ensures the lowest achievable concentrations in the leachate for many constituents. This in turn implies that potential long-term threats to this desired condition must be identified.
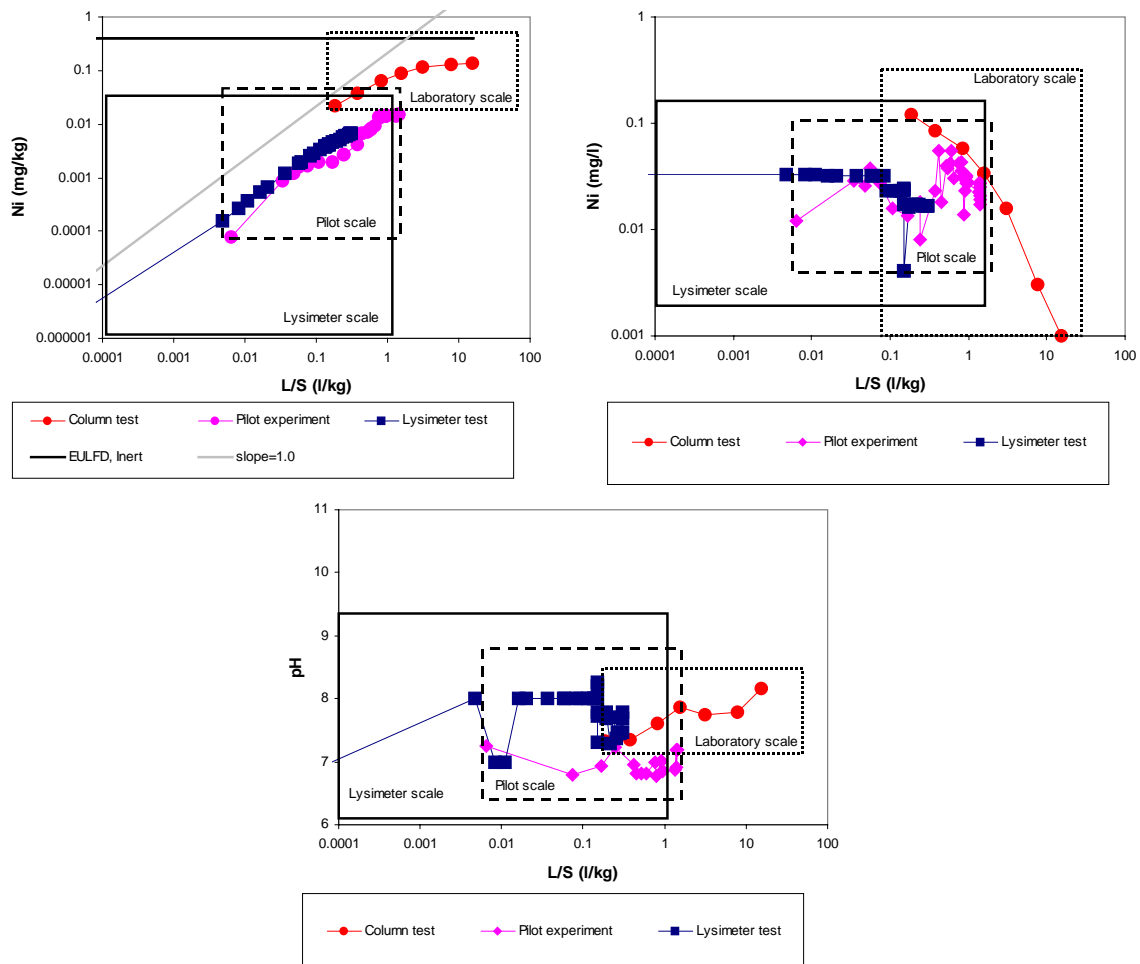
Figure 4   pH-dependent leaching of Ni at different scales illustrating that experimental scale provides information for extrapolating to different L/S ranges, as the trends in the data from different scales are consistent with each other.

Experiments on different scales were carried out in a framework of three full scale pilot projects to investigate different aspects of the theoretical approach. The first full scale pilot was set up to look into solving the issues arising for the organic waste landfill. The second full scale pilot was the Equifill concept for predominantly inorganic waste materials. The last pilot, targeted issues associated with the hazardous waste landfill, the Monolith. The latter concept was based on the cold immobilisation of the waste using cementing additives. The following paragraphs give a more detailed description of the experiments carried out in the three pilot projects.

3.2   Description of the Landgraaf bioreactor test cell

Basic design of the pilot

The pilot was constructed in 2001 as a small landfill cell with a rectangular footprint of 55x80 metres. The maximum depth of the waste is 9.5 metres with an average depth of approximately 6 metres. In order to optimise the infiltration and recirculation of leachate, two separate horizontal systems for leachate infiltration were installed. One horizontal system is situated at a depth more or less in the middle of the waste and a supplementary system is located below the top cover. Horizontal drains at the bottom collect the leachate. The test cell is isolated from the underlying landfill by a 2 mm HDPE (high density polyethylene) membrane, see Figure 5.
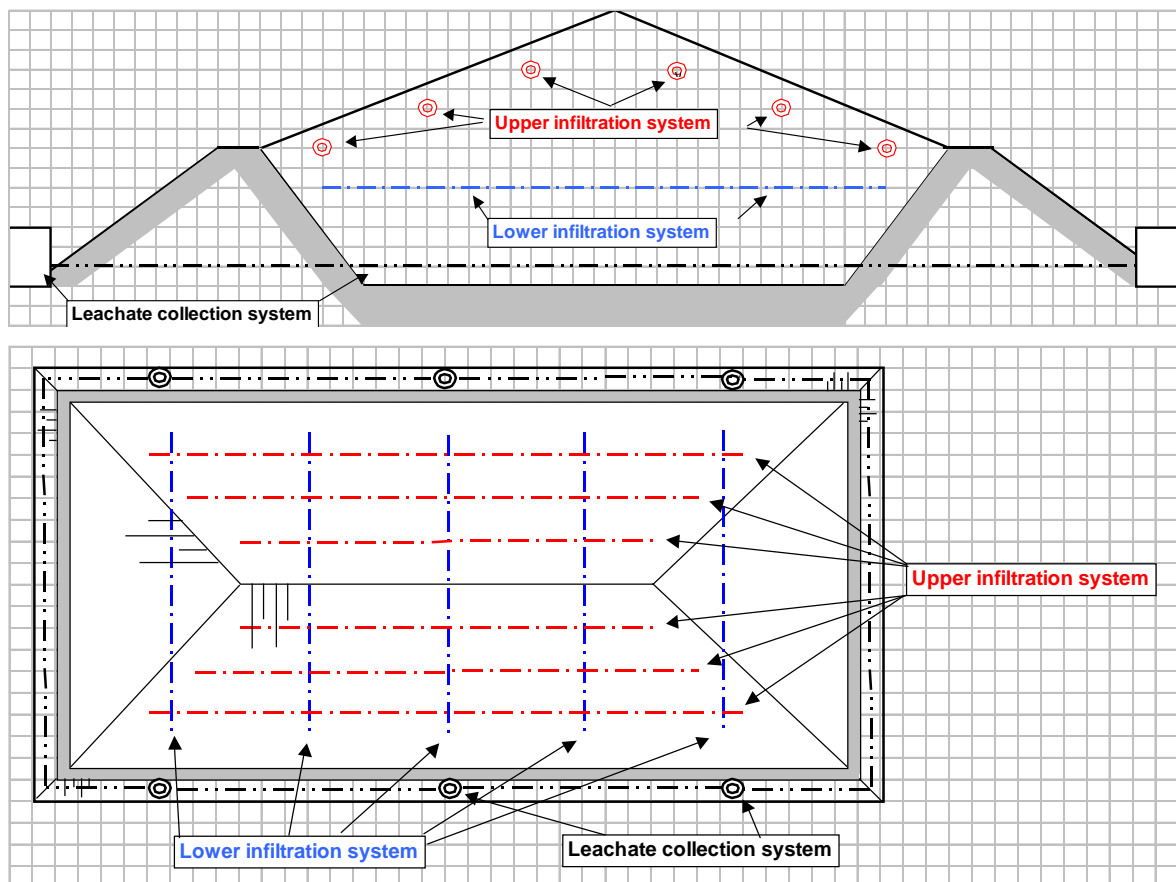
Figure 5   Design of bioreactor pilot

Landfill gas is extracted by six vertical wells and seven horizontal drains below the top cover. These drains lie in a bed of wood chips, and are covered with a geotextile and a thin layer of loamy sand. In addition to the gas extraction system, the construction of the top cover optimises the potential for methane oxidation. This semi-permeable top cover, existing of sandy soil enriched with coarse compost, allows the infiltration of some rainwater (*8*).

Characterisation of input

The organic waste is a mix of municipal solid waste and comparable industrial waste. The 48% of organic waste present in the mixture (Table 1) is not representative of the Dutch

situation but corresponds to landfill mixtures in southern and eastern EU countries and can be seen as a 'worst case' scenario for organic matter content.

Further waste characterisations were carried out in 2001 to achieve the required material composition (*8*). The waste characterisation determined the macro and micro parameters and the micro-organic pollution parameters. pH-stat experiments were also carried out. The results were used to compare the behaviour of the separate waste materials under the same circumstances.

The macro-parameters that were determined were e.g. inert salts (Cl^-), buffering components (Ca), biologically active components (Fe, DOC, N_{Kj} and SO_4^{2-}) and anions (F^- and B). The micro-parameters that were determined were e.g.: heavy metals (e.g. cadmium, zinc, copper, arsenic etc), volatile aromatic hydrocarbons, polycyclic aromatic hydrocarbons (PAH's), phenols and mineral oil.

As a result of these waste and composition studies the following waste materials were used in the pilot.

Table 1   *Quantities of waste-materials used in the pilot*

Waste-material	EWC code (*9*)	Weight-percentage (%)	Quantity (wet tonnes)	Quantity (dry solids)
Municipal Solid Waste	20.03.01	36	8,987	5,302
Industrial waste	20.03.01	12	2,988	1,404
Soil purification residue	19.12.09	19	4,718	3,762
Car shreddings	19.10.04	19	4,825	3,699
Moulding sand	10.09.08	5	1,335	1,322
Screening residues	19.12.09	8	1,928	1,600
Biological sludge	19.08.99	2	379	258
Total		100	25,160	17,348

Column experiments

The individual wastes were themselves subjected to pH-stat tests, at pH values of 4, 7, 11 and the natural pH of the material. To complete the picture, column-leaching tests were performed on mixed waste samples to determine the actual leaching characteristics, including the waste-waste interaction. Careful attention was placed on the production of representative samples for testing. The same material used in the leaching tests was also used in column experiments aimed at simulating the biological process. These tests were performed in two identical columns, each containing approximately 70 kg of mixed waste. No tests on the lysimeter scale were performed for the Landgraaf bioreactor.

Monitoring programme

Since the start of the pilot in 2002 all the water flows (infiltrated, recirculated, drainage as well as clean water supplements) and landfill gas extraction have been recorded on a daily basis. Once a week the quality of the leachate and infiltrate is analysed using several parameters, e.g. COD, NH_4^+, pH, EC (electrical conductivity) and temperature. Furthermore, the quality of the extracted gas is analysed and reported once a week. Periodically the quality of leachate, infiltrate, runoff (runoff water) and groundwater are analysed in a laboratory using a wide range of parameters. The results of these analyses are collected in a database that also contains the results of the other pilots conducted in the Netherlands (*4,10-13*).

Management of the different process phases

The reactor was constructed and filled with well defined waste in 2001. The recirculation of leachate and infiltration of water started in spring 2002. The operation of the bioreactor can roughly be distinguished in the following phases:

- Anaerobic degradation of the organic material enhanced by the recirculation of leachate.
- Flushing of salts and organic residues (DOC, humic and fulvic acids).
- Post degradation of microbial material and residual waste with special attention given to the removal of nitrogen by aeration of the landfill body and nitrification of the recirculated leachate.
- Stabilisation and humification of the residual waste with additional flushing to meet the Landfill Directive's standards.

In 2005 the third phase started: supplementary nitrification of the recirculated leachate using a submerged rotating biological contactor (RBC system). In 2006 the second element of this phase - alternating aeration of the landfill body via the water infiltration and discharge system - will start.

3.3   Description of the Nauerna Equifill pilot

Design of the predominantly inorganic waste landfill

The volume of the test cell is 12,000 m^3. The test cell is isolated from the rest of the landfill site by an 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. The runoff produced is collected separately and the amount measured. Once the test cell was completely filled, a layer of topsoil was used to cover the waste. No top liner was used in order to ensure percolation of the test cell.

Input of the predominantly inorganic waste landfill

The waste input into the pilot cell was controlled by more stringent acceptance criteria than currently required by legislation. The waste materials that have been landfilled in the test cell are described in Table 2. Samples were taken from all waste deposits landfilled in the cell and the landfilled weight of each deposit was recorded. Quality control measurements were taken on-site using short leaching tests. Every batch of waste was leached at an L/S ratio of 2 l/kg for one hour. The suspension was filtered (0.2 μm) and analysed for pH, electric conductivity, Cl^- and Dissolved Organic Carbon (DOC).

In the pilot predominantly inorganic mostly non-hazardous waste was accepted. Bulky wastes, which hardly contribute to leachate quality, can be accepted provided the normal rules of landfill stability are observed. Residual organic matter (non-degradable) in the waste is acceptable, and even desirable, as it binds metals and organic contaminants. Although it has limitations, a slowly degradable or non-degradable nature can be verified by a low DOC concentration. The target pH of the entire cell is 6.8 to 8. In order that this is maintained, the pH is measured and if it is < 5 or > 9 the acid (ANC) or base neutralisation capacity (BNC) is measured and balanced against the acid/ base neutralisation capacity of a mix of the main waste streams accepted in the landfill. The first layer of waste in a new cell must be a less critical waste (e.g. contaminated soil) to prevent initial leachate quality distortion.

Table 2   *Quantities of waste-materials used in the pilot. The moisture content of the total waste mixture is 36%*

EWC-code (*9*)	Type of waste	Tonnes	Percentage
	Non-hazardous waste		
01 05 08	Chloride containing drilling muds	467	2.7%
07 05 12	Sludges from on-site effluent treatment	15	0.1%
08 04 12	Adhesive and sealant sludges	3	0.0%
10 09 08	Casting cores and moulds	90	0.5%
12 01 17	Waste blasting material	7	0.0%
15 01 04	Metalic packaging	12	0.1%
16 03 06	Organic waste other than 16 03 05	26	0.2%
17 01 07	Mixtures of concrete, bricks, tiles and ceramics	53	0.3%
17 03 02	Bituminous mixtures other than 17 03 01	4	0.0%
17 05 04	Soil and stones other than 17 05 03	268	1.6%
17 09 04	Mixed construction and demolition wastes	55	0.3%
19 05 01	Non-composted fraction of MSW and similar	9	0.1%
19 10 04	Fluff light fraction and dust other than 19 10 03	21	0.1%
19 12 09	Soil and dredging sludge treatment residues	11,374	66.8%
19 12 12	Waste from mechanical treatment of waste	1,401	8.2%
19 13 02	Solid wastes from soil remediation	145	0.9%
20 03 03	Street cleaning residues	75	0.4%
	subtotal	14,025	82.4%
	Hazardous waste		
12 01 16*	Waste blasting material containing dangerous substances	289	1.7%
12 01 14*	Machining sludges containing dangerous materials	1,728	10.1%
17 05 03*	Soil and stones containing dangerous substances	950	5.6%
19 10 05*	Other fractions containing dangerous substances	36	0.2%
	subtotal	3,003	17.6%
	total	17,028	100.0%

Laboratory and lysimeter scale experiments

An integrated waste mix was prepared from all the waste samples collected by taking the waste mass per deposit into account. This waste mix was used for laboratory testing according to PrEN 14405 (column test; 0.0005 m^3) and PrEN 14429 (pH dependence leaching test), and for filling three lysimeters (1.5 m^3) (October 2001). Filling of the lysimeters was carried out as follows:

- Lysimeter 1: In order of delivery as practiced at the Nauerna pilot cell.
- Lysimeter 2: Encapsulation of the more contaminated wastes in relatively low permeability wastes.
- Lysimeter 3: In order of delivery as practiced at the Nauerna pilot cell with the addition of 5%$^{w/w}$ of sewage sludge and car shredder waste each, to increase organic matter loading.

The studies on the field, lysimeter and laboratory scales represent the different timescales (as a result of the different liquid to solid ratio at each scale) to which the waste was exposed. Laboratory leaching tests reflect the long-term leaching behaviour of the waste material, with

their large L/S ratio range (0 to10 l/kg). The lysimeter experiments and the field study reflect the actual leaching behaviour. The lysimeter study might develop a slightly higher L/S ratio as a function of time due to its relatively small scale. The data from the different scales of testing was compiled in order to address the laboratory-field relationship of contaminants.

Monitoring programme

Monitoring of leachate and porous ceramic sampling cups (most dry) started in 2001 and continues at a frequency of 6 to12 samples per year. The leachate from the lysimeters and pilot, and the extracts from laboratory tests are analysed for major, minor and trace elements (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) by ICP-AES (inductively coupled plasma-atomic emission spectrometry) DOC, COD, Cl^-, SO_4^{2-}, F^-, NH_4^+ and NO_3^-. Together with information on test performance, pH, conductivity, redox and water volumes the data are stored in the database in a unified data format (see section 3.9).

Management of the different process phases

The filling of the pilot (12,000 m^3) started in April 2000 and was completed in November 2001 when a soil top cover was placed over it, see Figure 6. If the waste can be considered inert, less stringent top cover requirements could be applied than would be required by the current Dutch regulation.

Figure 6   Design of Equifill pilot

3.4   Description of the Monolith pilot

Design of the stabilised waste Monolith landfill

Many complex physical and chemical processes control the releases from a stabilised waste monofill. Therefore, the first goal is to understand the physical and chemical processes occurring in a stabilised waste monofill. Important processes for a stabilised waste landfill are

outlined in Figure 7. Vertical drainage channels in the landfill are created instead of creating one large monolith. A soil drainage layer is also placed underneath the stabilised waste. This soil layer buffers the alkaline percolate water and might also function as an adsorption layer for heavy metals (onto Fe/Al-oxides and solid organic matter). The pilot project consists of four test cells (A, B, C and D) with varying waste recipes and cell heights.
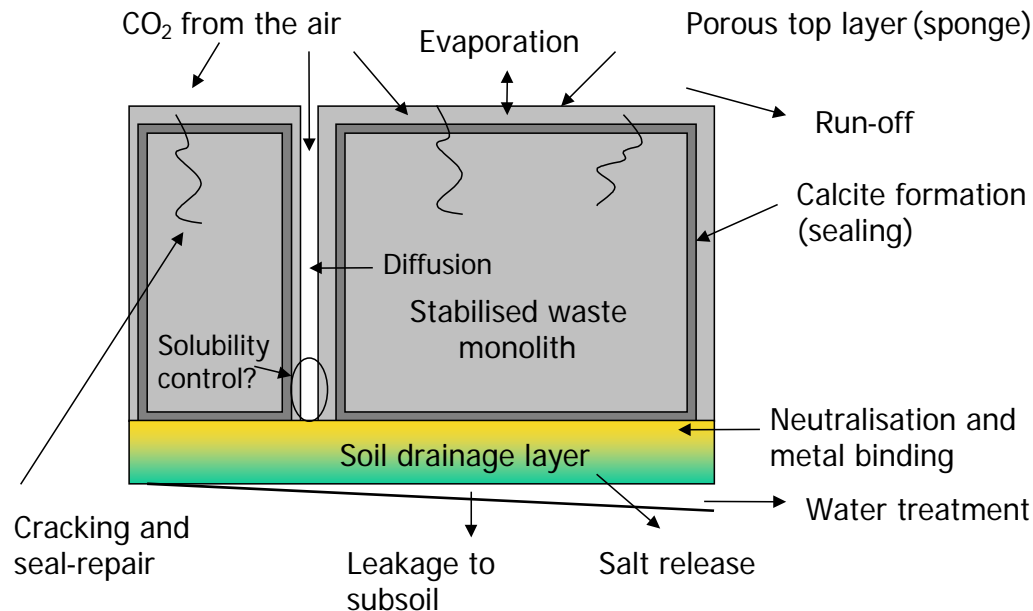

Figure 7   Schematic representation of a stabilised waste Monolith landfill with important physical and chemical factors that can influence the environmental behaviour.

At the site of VBM (Maasvlakte, The Netherlands) a pilot study of stabilised waste has been realised. A cross section of the pilot scale monofill is shown in Figure 8. The pilot monofill is divided into four sections in which the solidified/stabilised waste is landfilled and the effects of height, carbonation and mixing of recipes studied. The waste is stabilised *in situ* in layers of approximately 0.5 m and is separated at intervals of 1.5 m by vertically placed geotextile. The space filled by the geotextile is to create preferential flow channels for water, instead of one very large block, as part of the sustainable concept for stabilised/solidified material. This reduces the amount of water percolating through the stabilised waste to avoid saturation.

Facilities are installed to allow sampling of the different types of water from the different sections. For each section three types of percolate are distinguished: pore water or permeation water, leachate and runoff. In the experiment pore water is the displaced water that flows through the blocks. The pore water is sampled by placing a plastic foil on a slight slope in the drainage sand directly beneath the blocks. The pore water is collected in a polyethylene container and can be sampled with a porous ceramic cup: precautions are taken to prevent CO_2 poisoning from the air. In the experiment, leachate is regarded as the water that preferentially flows through the spaces between the blocks. Runoff is the water that does not infiltrate into the blocks and that flows across the surface of the blocks. The runoff is sampled from each section via a gutter at the edges of the blocks.

Stabilised waste cores were drilled from the top layer of the pilot experiment to assess the leaching behaviour as a function of depth in the compartment. The drilled cores were

transported to the laboratory, sliced and leached at the natural pH of the suspension (L/S=10). Analyses of heavy metals and oxyanions were undertaken.

At the bottom of the stabilised waste monofill, a slightly contaminated soil was used as a stabilisation layer to protect the liner system. This has proved to be a very useful means of controlling the leachate quality from the stabilised waste monofill. The pH buffering and the retention of metals by the soil layer lead to a leachate with a neutral pH and rather low metal concentrations. A key question now is how more effective use can be made of the behaviour of the soil layer. By increasing the buffering potential of the soil layer beyond the period of direct infiltration, a favourable leachate quality can be maintained over much longer time scales. From a pH-dependent leaching test, the buffering capacity of the soil used can be determined and balanced against the infiltration and carbonation of the stabilised waste.
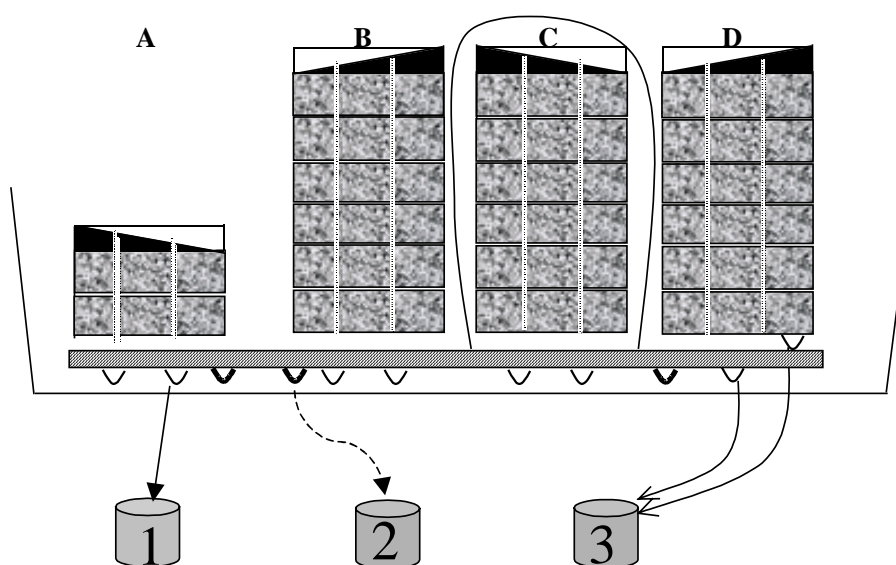

Figure 8   Set-up for pilot scale monofill of stabilised/solidified hazardous waste. The width of the each cell is 4.5 m with a vertical layer of geotextile at each 1.5 m. The first cell (A) is 1 m high, cells B to D are 3 m high. Sampling facilities have been made to collect percolate water and runoff.

Table 3   *Quantities of waste-materials used in cell D of the pilot. Cells A (51 tonnes wet weight), B and C (102 tonnes wet weight each) were constructed with a stabilised MSWI fly ash recipe. EWC codes are given in parenthesis.*

Waste-material recipe	Weight-percentage (%)	Quantity (wet tonnes)
APC residue (wet system, 19.01.07*, 11.01.09*, 19.01.05*) and MSWI fly ash (19.01.13*)	50	51
Metal sludge (11.01.09*) and MSWI fly ash (19.01.13*)	25	25.5
MSWI fly ash (19.01.13*)	17	17
MSWI filter cake (11.01.09*)	8	8.5
Total	100	105

Input of the stabilised waste Monolith landfill

Cells A, B and C were all filled using one stabilised waste recipe (see Table 3), which contained MSWI fly ash. Cell D contains a mixture of recipes; each contained different waste

materials. Table 3 shows the proportion of each waste material and their respective wet weights used in the stabilisation plant for cell D. The quantities mentioned reflect the weights of the waste, binder material (different amounts of binder are used for the various waste materials) and any water added. Other waste materials that are commonly stabilised in practice are: MSWI fly ash, filter ashes, A-cokes, metal sludges and filter cake from wastewater treatment plants.

Monitoring programme

Stabilised waste samples were taken from the full scale pilot just after all the materials were mixed. They were cured for at least 28 days before testing. Some 14 different stabilised waste recipes were characterised by pH-dependent leaching and tank leaching tests. During the pilot the percolate and runoff containers were emptied regularly. The water volumes, pH, EC and temperature were measured on-site. Samples for chemical analysis were taken about every two months. Analyses were performed on filtered samples (0.45 μm).

Management phases

The pilot was constructed and filled with well defined stabilisation recipes as indicated in Table 3. Construction of the pilot started in February 2003 and was completed in July 2003. The monitoring programme started in September 2003 and will continue until the spring of 2006. After this phase, part of the pilot will be stopped and the research will focus on the processes that have occurred in the stabilised waste during the previous three years. The cells will be destructed so that samples from several places within the waste body can be taken, namely: upper layer, material near the vertical drainage and material at the bottom of the compartment. In addition, the soil layer beneath the waste material will be sampled and analysed in the laboratory. This will allow conclusions to be made on the progression of carbonation within the stabilised waste drainage channels. An impression of the overall structure and stability of the stabilised waste mass will also be gained. Furthermore, it will provide an insight into the performance of the soil layer in buffering alkalinity and binding the metals released from the waste. This information will be used to outline design characteristics for a sustainable stabilised waste landfill in terms of the soil layer thickness and its properties, and the need to stimulate carbonation during a given period.

3.5   Approach to modelling landfill processes

Modelling the processes occurring in landfills which influence the leaching of contaminants requires that a large number of chemical, biochemical and transport phenomena are taken into account. These are namely:
1 Biochemical conversion of organic materials into dissolved organic materials and the subsequent formation of fatty acids, N_{Kj} and biogas.
2 Physical chemical speciation involving solution and precipitation of metals, anions and salts; and the adsorption of components.
3 Transport of liquids through pores in the waste.
4 Transport of gases that are formed through pores in the waste.
5 Generation and transport of heat that is generated as a result of aerobic and anaerobic reactions within the waste.

The objective of the modelling activities within the framework of this study is to predict (i) the pollution potential of the landfill in the long-term to extreme long-term (from decades after deposition of the waste into eternity) and (ii) the effects of pollution control measures used in this study on this pollution potential. This is a very specific modelling objective and it allows a

significant simplification of processes, compared to landfill models that are developed for more general purposes.

The approach used and developed in this project is based on the integration of the first three processes (sub-models) listed above: (1) biochemical conversion, (2) physical chemical speciation, and (3) transport of liquids are combined to give predictions on the concentrations of organic components, N_{Kj} and all major and minor elements over time, see Figure 9. The sub-models are also simplified, e.g. the organic components in the model are divided into three classes: humic acids, fulvic acids, and fatty acids. This does not reflect the fact that the situation is more complicated in reality, but for the prediction BOD, COD and trace metals levels in the leachate over long time periods, more detail is not required.
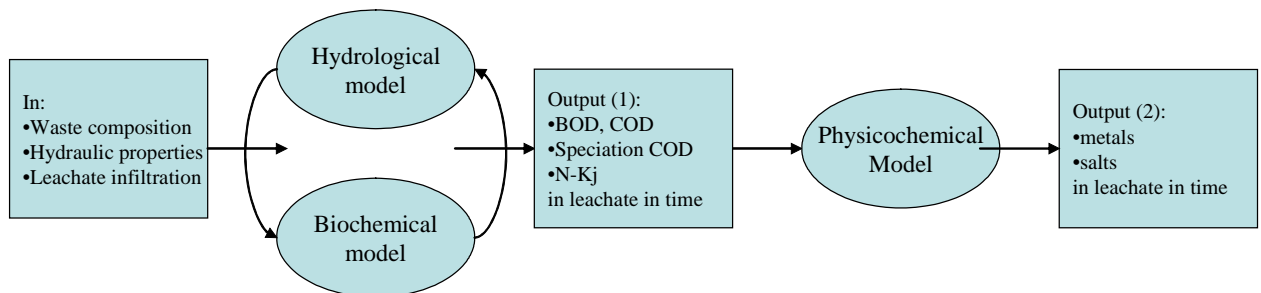

Figure 9   Biochemical and hydrological approach

Two of the phenomena listed above are not taken into account, as they are either considered to be of minor importance, or their effect can be simplified and taken into account in an alternative way:
- Gas transport (4) is not taken into consideration. It only occurs in the first few years and does not influence the hydraulics in the long-term. The main effect of gas transport in the initial phases is to reduce the hydraulic permeability of the waste, as gas-filled pores are not available for liquid transport and the effective porosity is thus reduced. The effect of gas transport can be taken into account by assuming a reduced hydraulic permeability in the first few years after depositing the waste.
- Generation and transport of heat (5) is not modelled either, as little heat is produced under anaerobic conditions, so its effect is small. The only effect it has is on the rate of bioconversions and this can be neglected for two reasons: (a) its impact is small compared to e.g. the impact of leachate recirculation; (b) the speed of bioconversion (within limits) is not a major determining factor for the long-term leaching behaviour of the landfill. The validity of the latter assumption is demonstrated by the modelling results.

3.6   Modelling biochemical transformation

The levels of BOD, COD and N_{Kj} in leachate are of major importance within landfills with substantial amounts of organic carbon, Ultimately BOD and COD in landfill leachate have to meet certain criteria. Although no leaching limit values are defined for N_{Kj} in the Landfill Directive's acceptance criteria, given in Annex II (2). N_{Kj} is a component of importance because of its potential environmental impact and so cannot be ignored in this project. The model in the form used for the calculations does not include the direct effects of aeration or the indirect effects of aerobic reactions resulting from the introduction of nitrate in the anaerobic environment of the landfill. It has been found that COD correlates with DOC in case of the bioreactor experiments. High levels of DOC lead to increased concentrations of heavy metals

in leachate due to complexation. DOC has to meet certain criteria in order that the criteria for heavy metal concentrations in leachate are met.

In order to understand how biochemical degradation influences the leaching of organic components and N_{Kj}, a model (see Figure 10) has been developed. It is based on an elaboration of the reaction scheme proposed by McDougal and Philp (*14*). In this reaction scheme, several steps are distinguished, namely:

- Hydrolysis: the solid organic fraction (SOF) is broken down to a complex mixture of organic and inorganic components that in turn are converted into other (smaller) dissolved organic components. In this model, the speciation of components is limited to three classes in order to serve as inputs into the physicochemical model: fulvic acids, humic acids and volatile fatty acids (VFA).
- Methanogenesis: Ultimately VFA is converted further into biogas. During generation of biogas, methanogenic biomass (MB) is formed. MB is the bacteria responsible for biogas production.
- Death of methanogenic biomass.
- Decay of dead biomass, releasing N_{Kj} again and a small amount of organic carbon, and conversion of the organic carbon as a result of release by the dying methanogenic biomass.
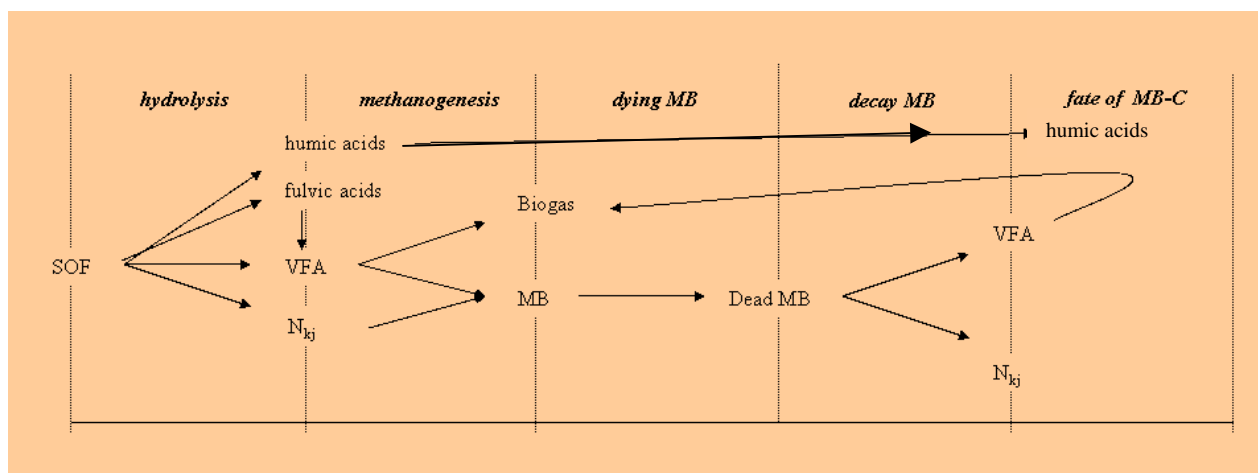

Figure 10   Schematic overview of the biochemical modelling of leachate quality development

In reality the reaction mechanisms are understood to be much more complex. In this simple model, VFA represents all readily degradable material, fulvic acids represent all slowly degradable dissolved organic material, and humic acids represent all non-degradable dissolved organic material. The MB describes all bacteriological biomass, using organic material and nutrients. It is also a pool of material that prolongs its own lifetime by growing on its residues. The reaction scheme was used within the Orchestra modelling framework developed by Meeussen (*15*).

3.7 Physical chemical speciation modelling

Physical chemical speciation modelling was also used within the Orchestra framework developed by Meeussen (*15*). Aqueous speciation and selected mineral precipitation reactions were taken from the MINTEQA2 database. Ion adsorption onto dissolved and particulate organic matter were calculated using the NICA-Donnan model (*16*), together with the generic

adsorption reactions as published by Milne *et al.* (*17*). Adsorption of ions onto iron and aluminium oxides were modelled according to the generalised two layer model of Dzombak and Morel (*18*).

The input into the model of metal availabilities, selected possible solubility controlling minerals, active Fe (hydrous ferrous oxide (HFO)) and Al-oxide sites (Fe- and Al-oxides) were summed and used as an input for HFO as described in (*19*)). Particulate organic matter and a description of the DOC concentration as a function of pH (TS 14429, 2005) were also input into the model. Essentially, the speciation of all the elements with the same parameter settings is calculated in one 'problem definition' for use in the model, to obtain a 'forward' model prediction. This limits the degree of freedom in selecting parameter settings considerably, as an improvement of the model description for one element may deteriorate the outcome for other elements. As a starting point for the model calculations, the maximum value as obtained in the pH-dependent leaching test (between pH 3 and 13) was used as the fraction relevant for environmental impact modelling (availability).

The mineral phases that were allowed to precipitate were selected after calculation of their respective Saturation Indices (SI) in the original pH-dependent leaching test eluates. Saturation indices were calculated for all > 650 minerals in the thermodynamic database and a selection of the most likely and relevant phases was made based on (i) the likeliness of their existence in waste systems and (ii) the degree of fit over a wider pH range and (iii) the closeness of the log SI value to 0 (the theoretical SI value for saturation).

The output of the speciation calculations consists of (i) the predicted total amounts in the water phase and (ii) the calculated speciation of each of the elements in the different phases and reactive surfaces considered in the model. Based on the adequate geochemical model descriptions of the pH-dependent leaching data, the chemical speciation/the partitioning in both the solid phase and the leachate can be calculated, more information is available in detailed research reports Equifill (*20*) and Database & Modelling (*21*), which are also referenced in section 6.3.

3.8   Modelling hydrology

Water flow modelling in waste was also done within the Orchestra framework. It was assumed that water flow takes place within a triple porosity concept in which the pore volume is divided between a stagnant bulk, a slow mobile phase and sharp preferential channels (a fast mobile phase through which most of the fluid movement takes place). Exchange of water-dissolved compounds between the stagnant bulk to the mobile phases takes place through diffusion. The slow mobile phase is conceptually situated between the stagnant bulk and the fast mobile phase. Flow in the mobile phases is described by a convective dispersive approach. Flow rates are not explicitly simulated but are boundary driven. Figure 11 is a schematic representation of the hydrological model.

The model was tested using the results of bioreactor column tests and was subsequently used for predicting the behaviour of the Landgraaf test cell.
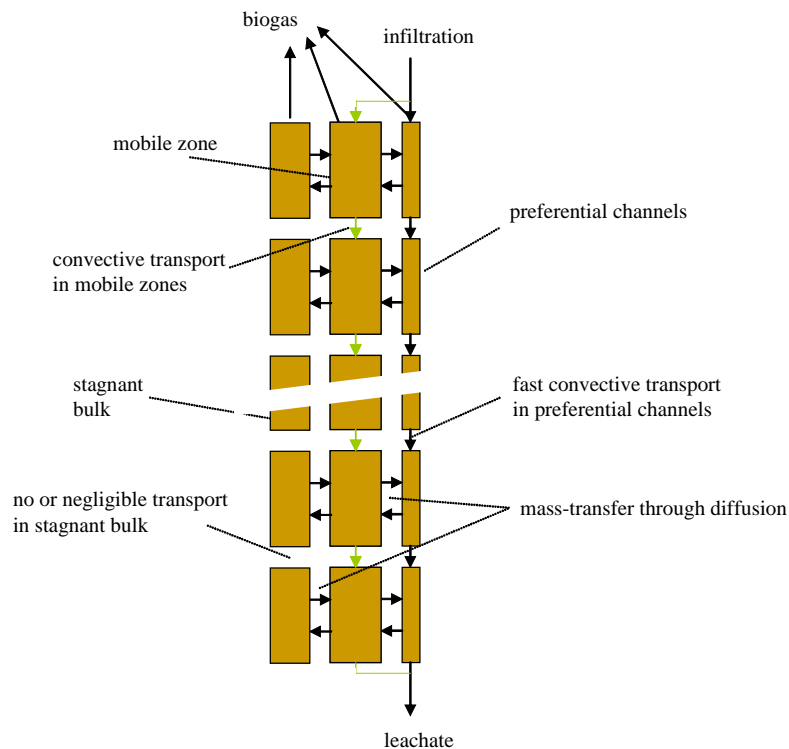
Figure 11   Schematic representation of the hydrological flow in the landfill

3.9   **Data interpretation and modelling framework: LEACH-XS**

In order to efficiently cope with the very large amount of data collected in the experiments
carried out on the different scales, the sophisticated database/expert system LeachXS was
used (*13*). Figure 12 is an overview of the LeachXS database/expert system. The core of the
system is a relational database that allows the storage of all the types of raw experimental
data collected during this project and many others. The database contains data on
(contaminated) soils, sludges, sediments, composts, stabilised wastes, mining wastes, treated
wood, and a wide range of construction materials obtained from this project, from the literature
as well as digital data from other groups and experiments carried out on a wide range of
materials in different contexts (*4,10,13,22*). Information stored in the database includes the
type of sample, sampling location, originating landfill, type of experiment, general sample
characteristics and analytical results. An important characteristic of the database is that all the
raw data are stored, e.g. for a pH-stat experiment, all the concentrations measured as a
function of pH are stored for a particular sample. Raw data from other experiments carried out
on the same sample are also stored, including the results of field measurements recorded at
the time of sampling.

The second important feature of the LeachXS database/expert system is the user interface.
The user interface makes it very easy to present similar data in charts from a very wide range
of samples covering a wide range of conditions.

The third important feature of LeachXS is the fact that expert functionality is continuously
added to the system. Using the Orchestra framework developed by Meeussen (*15*),
geochemical equilibrium modelling can be performed for samples that have been completely
characterised.
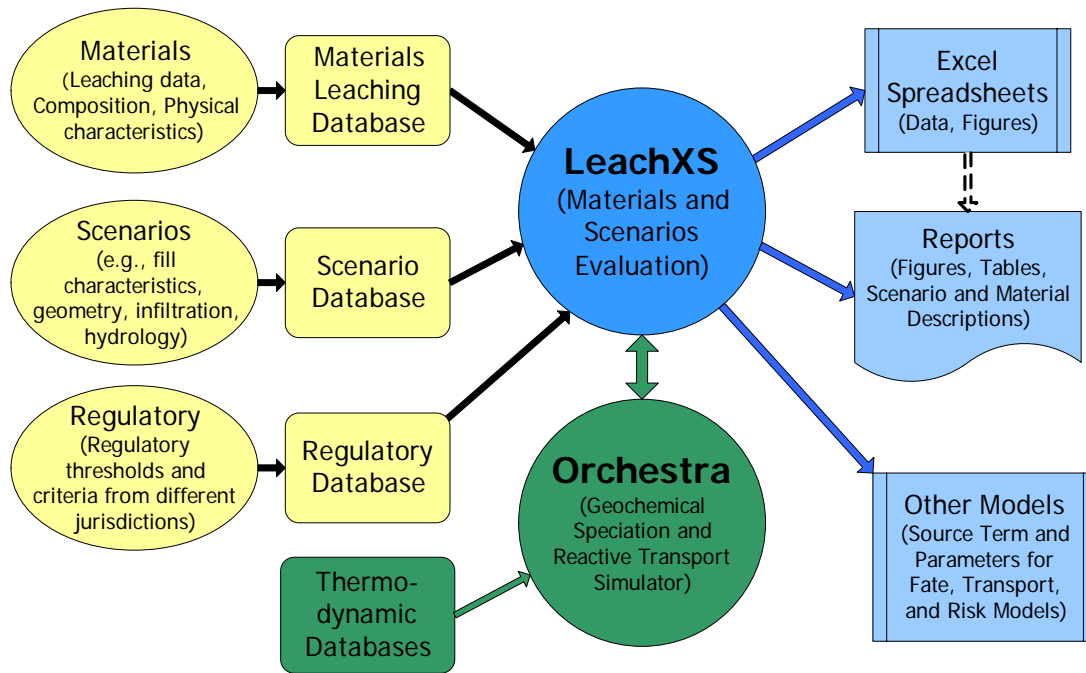
Figure 12   Structure of LeachXS

Using the Orchestra framework (*15*), solute transport modelling functionality, with a multiple porosity approach (fast mobile, slow mobile and immobile zones), is also included in LeachXS. This latter functionality allows the user to simulate leaching experiments and extrapolate the results to other leaching situations. As explained in previous paragraphs, the solute transport model has the capacity to simulate both the inorganic equilibrium geochemistry and the associated degradation of organic matter.

Another form of expert functionality is a direct comparison of leaching test results for a certain material with the relevant regulatory standards.

4.   Results & Discussion

4.1   General results

Influence of waste properties on leaching behaviour

The results from geochemical modelling show that there is an understanding of the chemical processes that determine the leaching behaviour of a significant number of elements from the waste mixture. This information formed a basis for sensitivity analysis on the influences of changing contaminant availability, amounts of organic matter and/or HFO in the model. Such a sensitivity analysis should be a first step towards certain waste management decisions, e.g. for landfill owners.

Two examples have been worked through in detail below.

Prediction of pH-dependent leaching behaviour
Interpretation of results

To facilitate the interpretation of the measured and predicted leaching behaviour, an example is given Figure 13. The leaching data is represented as a function of pH by the red data points. The black solid line represents the predicted total concentration of the considered element in solution, which should ideally meet the data points to show that there is a good understanding of the chemical processes that determine the leaching behaviour. Furthermore, Figure 13
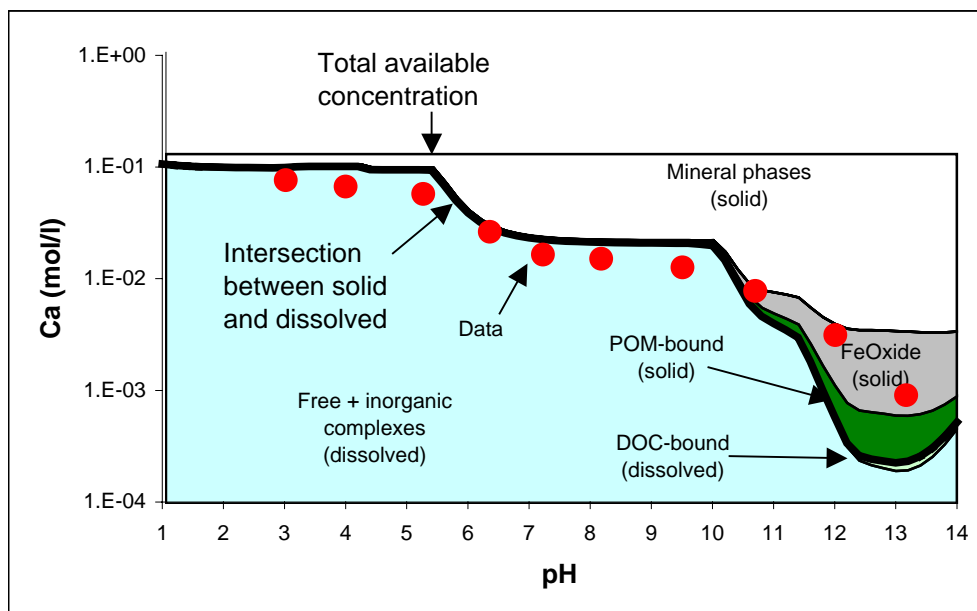
Figure 13   Example of integrated data presentation for pH-static leaching test results and geochemical speciation modelling. Red data points represent leaching data and the black solid line is the predicted leached concentration. Areas represent the element speciation: White = minerals, Grey = Fe-oxide sorption, Dark green = complexation to solid organic carbon, Light green = complexation to dissolved organic carbon and Light blue = free and inorganically complexed form (*23*).

shows the calculated chemical speciation of the element in both the solid matrix and the sample solution. The predicted leaching behaviour is therefore the intersection between the calculated speciation in the solid matrix (minerals, sorption to Fe-oxides and binding to solid organic matter) and in solution (free, inorganic and complexed by dissolved organic carbon). This presentational form integrates the predicted total leached concentration and the different species that determine the leached concentrations.

The upper line in Figure 13 represents the total available concentration (input into model). The white area shows the amount of the element bound as minerals in the solid phase. The grey area represents sorption onto Fe-oxides, whilst complexation to solid organic matter is presented in dark green. These areas represent the total amount in the solid matrix as a function of pH. The light blue area is the total amount of the free ion and the inorganically complexed form in solution. The light green area represents the amount of the element that is organically complexed.

The first example deals with the calculated Pb emissions in the waste mixture as a result of an increased Pb availability (see Figure 14). The initial calculations are given in Figure 14A. It can be seen that Pb is substantially bound to HFO in the solid phase at neutral to slightly acidic pH values. Figure 14B shows the measured and predicted pH-dependent leaching behaviour of Pb when the availability of Pb was increased by a factor 10. Figure 14C shows the situation when Pb availability was increased 10 fold and the DOC concentration was increased by a factor of 3 in the model. It can be seen that the predicted Pb emissions in the neutral to acidic pH region significantly increased as a result of the increased availability. This implies that increasing Pb availability (e.g. by accepting a high Pb-containing waste in to a landfill), when other waste mixture conditions are unchanged, will directly result in increasing Pb emissions, and vice versa. It can be seen that Pb emissions increase mainly as a result of complexation with organic matter (Figure 14C). An increase in both the Pb availability and the HFO content of the waste mixture leads to a subsequent reduction in Pb emissions (Figure 14D). Only at pH values lower than 6, is an increase in emissions observed, as shown in the initial calculations (Figure 14A). Binding to HFO is not the dominant solubility control mechanism in these conditions. However, these low pH values will probably not be relevant in practice.

The third example of the sensitivity analysis is given for the leaching of Zn (Figure 15). The initial model results are given in Figure 15A and show that Zn is primarily in the mineral phases at neutral to alkaline pH values. The Zn leachability was calculated for an availability 10 times higher without changing any of the other parameters. Figure 15B shows that there is a limited effect on Zn leaching in the neutral to alkaline pH range when availability is increased to a factor 10. The solubility of the mineral Willemite determines the dissolved Zn concentrations. Zn concentrations are not dependent on the total amount of (available) Zn in the system. However, Zn leaching increases at very low pH values because the available Zn concentration is higher (Zn becomes very soluble in the acidic pH domain).

Increasing DOC concentrations has an effect on Zn leaching (Figure 15C) but not to the same extent as shown for Pb. The increased HFO content does not effect the leached Zn concentrations (Figure 15D). However, it can be seen that speciation in the solid phase changes in the neutral pH region. Sorption onto HFO is a more important process in the binding of Zn to the solid matrix.
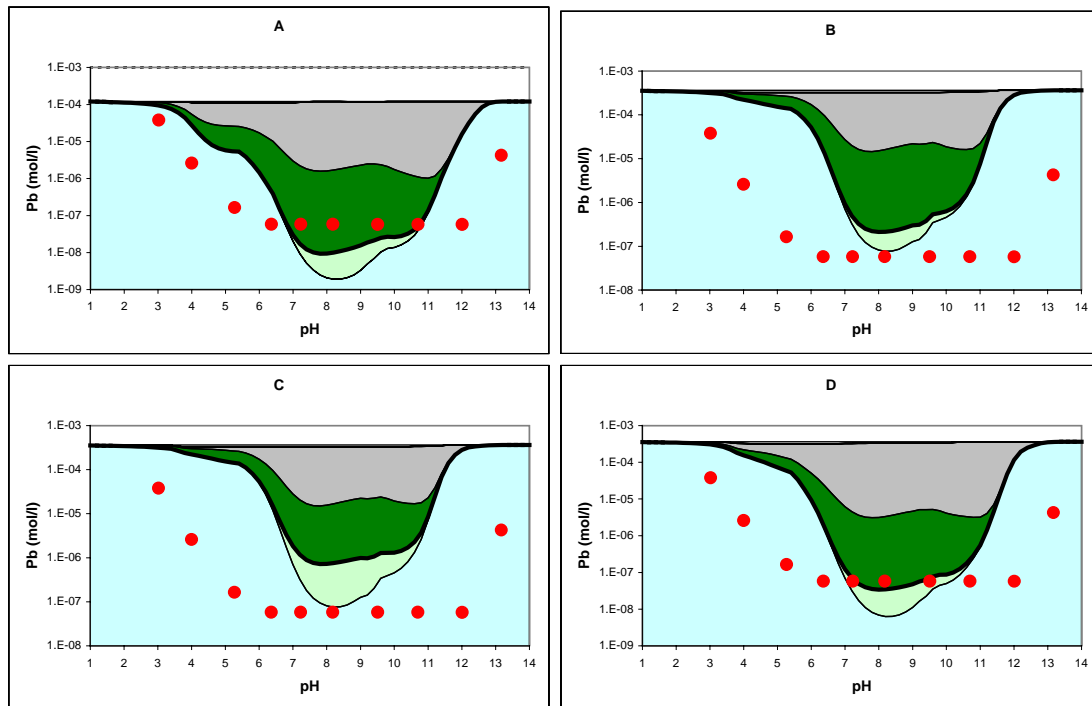
Figure 14   Prediction of Pb leaching as a function of pH from a predominantly inorganic
             waste mixture under varying conditions. A: Initial calculations; B: Pb availability is
             increased by a factor of 3; C: Pb availability and DOC concentration are increased
             by a factor of 3; D: Pb availability and the content of HFO are increased by a
             factor of 3 (*23*).

Influence of increased availability on the leaching behaviour of other elements

The results presented in Figure 14 and Figure 15 show that the leaching behaviour of
contaminants can be adjusted by changing the properties of the waste mixture. Therefore,
there are opportunities to improve waste management options by the introduction of
tolerances in the available contaminant concentrations. This can be practiced by controlling
the total available amounts of contaminants relative to the amounts of HFO and solid organic
matter. However, increasing availabilities for one metal might affect the leaching behaviour of
other elements due to competition for binding sites on HFO and solid organic matter, or
mineral formation.

Figure 16 shows the effect of increased Pb availability (factor 10) on the leaching behaviour of
Mo and V. As mentioned previously, Pb influences the leachability of Mo and V through the
formation of Pb-molybdate and Pb-vanadates. Increasing the Pb availability changes the
predicted Mo and V leachate concentrations significantly. In this example, the predicted
leaching behaviour of Mo and V is substantially lower as a result of an increased Pb
availability. However, changing contaminant availabilities could also result in enhanced
leaching of other contaminants due to competition on surfaces (HFO and/or organic matter).
Consideration of these results emphasises the need to ensure that the improvement of the
environmental quality of waste materials must be assessed by studies which include
measurements and modelling of all the relevant elements/compounds.
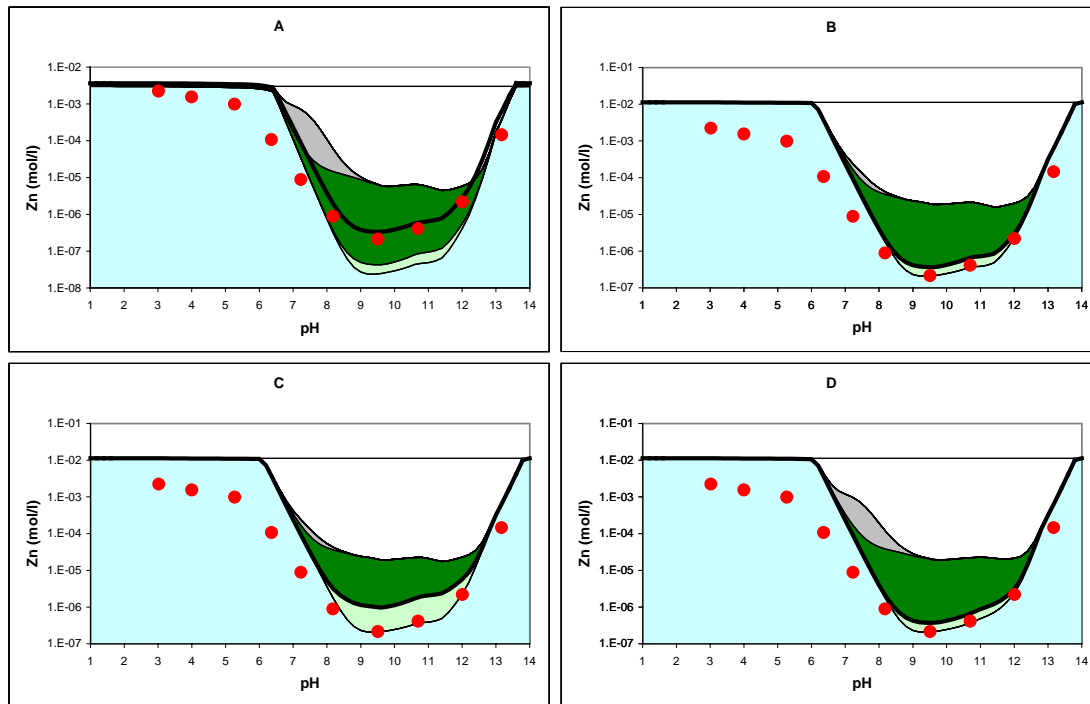
Figure 15   Prediction of Zn leaching as a function of pH from a predominantly inorganic
            waste mixture under varying conditions. A: Initial calculations; B: Zn availability is
            increased by a factor of 3; C: Zn availability and DOC concentration are increased
            by a factor of 3; D: Zn availability and the content of HFO are increased by a
            factor of 3 (*23*).

Figure 16   Prediction of the leaching behaviour of Mo and V as a result of an increased Pb
            availability. Figure A and B shows the initial prediction for Mo and V respectively.
            Mo (C) and V (D) leachability after increasing Pb availability by a factor of 10 (*23*).

Influence of reducing conditions on the leaching behaviour of Cu, Fe and Mn

Reducing conditions are dominant in a landfill scenario for a significant amount of time. However, laboratory leaching tests are generally performed under atmo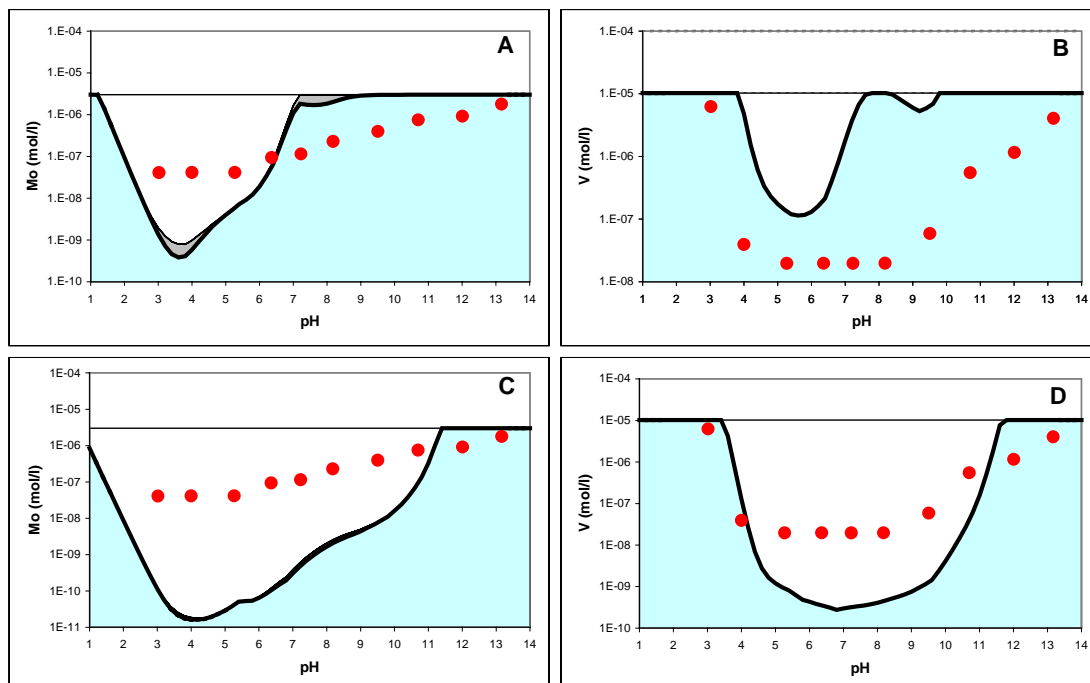spheric conditions. This aspect needs to be taken into account in the estimation of the long-term prediction of contaminant emissions. Reducing conditions have a pronounced effect on the leaching behaviour of waste materials in a landfill.

Figure 17 shows the change in leachability of Cu, Fe and Mn after imposing reducing conditions on the system (pe + pH = 6). Cu leachability is significantly reduced after imposing reducing conditions. In the case of Fe, the solubility curve shifts to a higher pH as the more mobile Fe(II) is formed. For Mn, increased mobility is observed and can be explained by the formation of the more mobile Mn(II).

More information on the research on modelling is presented in the research report Database & Modelling (*21*), which is also referenced in section 6.3.
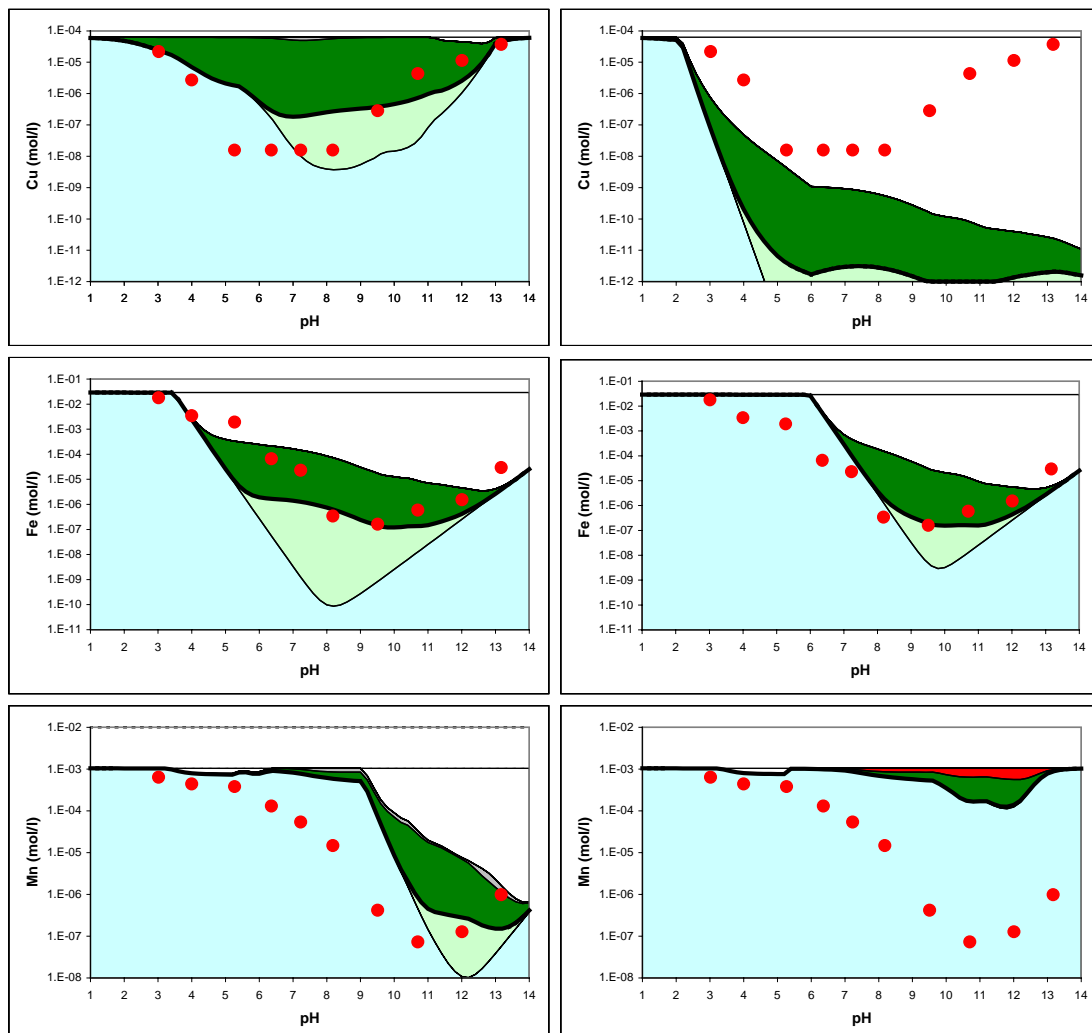
Figure 17   Prediction of Cu, Fe and Mn leaching behaviour in a predominantly inorganic waste mixture as a function of pH. The left graphs show the initial modelling results, the right graphs show the results under the assumption of reducing conditions (*23*).

4.2   Bioreactor: organic domination

Water management and leachate quality

Water management can be characterised by the 'fill and draw' principle. During the 'fill' period the leachate is recirculated. Recirculation and the addition of fresh water achieve a 3,000 mm per year infiltration rate. In this period anaerobic degradation is stimulated and soluble components increase in the water system.

During the extended 'draw' period the soluble components are drained out of the landfill body. At first the leachate in the mobile zone is drained at the end of the draw period and is supplemented by leachate (and diffused soluble material) from the semi-stagnant zones in the landfill body. The successive periods of 'fill' and 'draw' are represented in Figure 18. It can be seen that after initial build up in the first year, the difference between fresh water input and drained volume remains more or less constant, indicating saturation in the part of the pilot that is reached by the infiltration system.
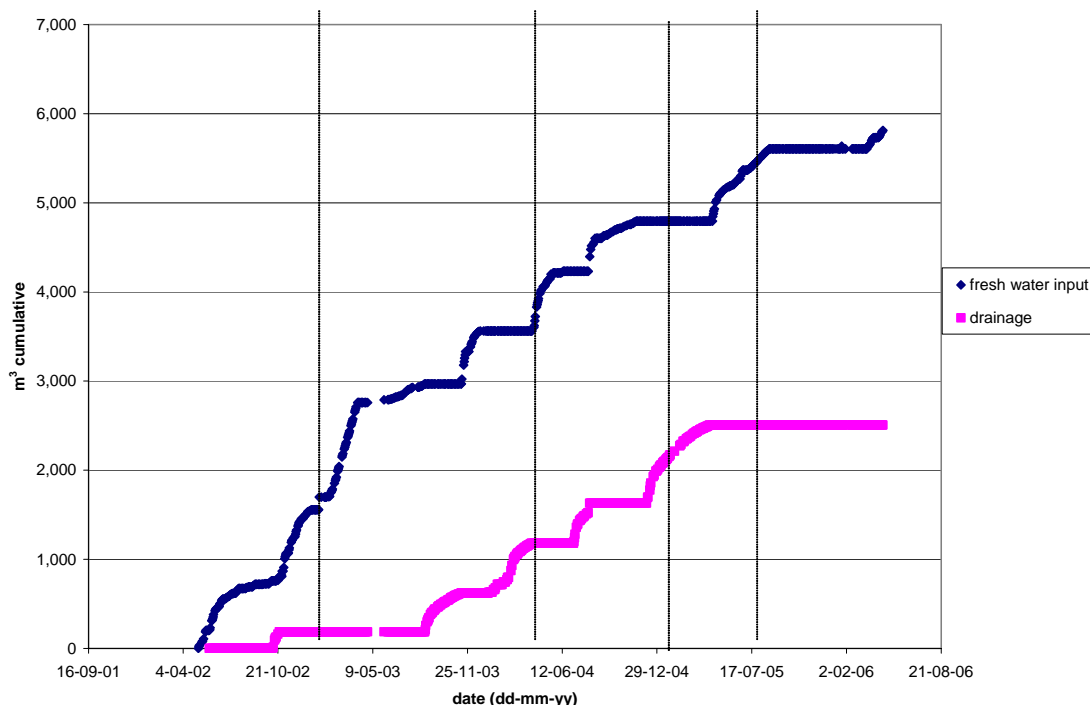
Figure 18   Fill (fresh water) and draw of the bioreactor

The sodium leached in the draw period of the bioreactor ('pilot') was compared with the column leaching tests ('ECN', laboratory scale) and presented in Figure 19. It can be concluded that laboratory scale leaching and pilot leaching are very similar. The gradient of the line for the pilot is however only 70% of that of the laboratory scale tests. This indicates that some of the material in the pilot was not (yet) participating in the leaching process.

The influences of anaerobic degradation, together with the supply of fresh water (and recirculation) and the drainage of leachate are shown in Figure 20. The sharp decrease in the level of COD was due to the supply of fresh water. The sharp increase in COD levels was caused by the drainage of semi-stagnant leachate at the end of the draw period. An overall decrease in the COD level is shown and can be attributed to the practical completion of the anaerobic degradation.
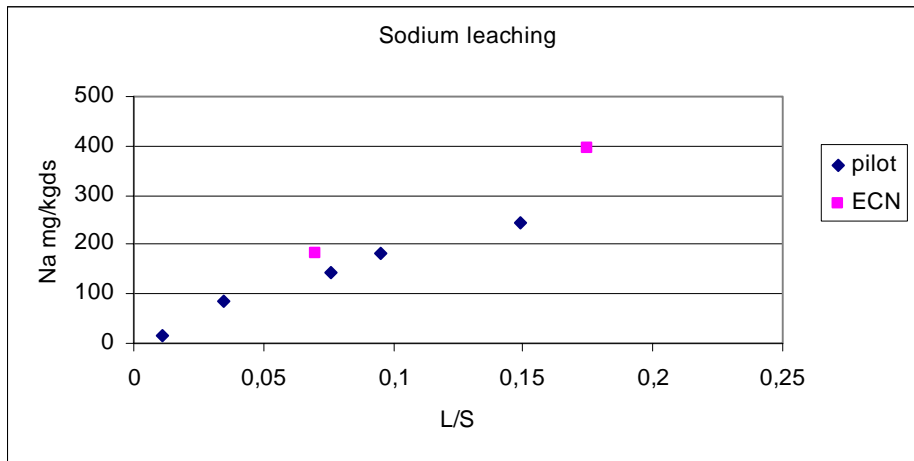
Figure 19   Sodium leaching on laboratory scale (ECN) and pilot scale.
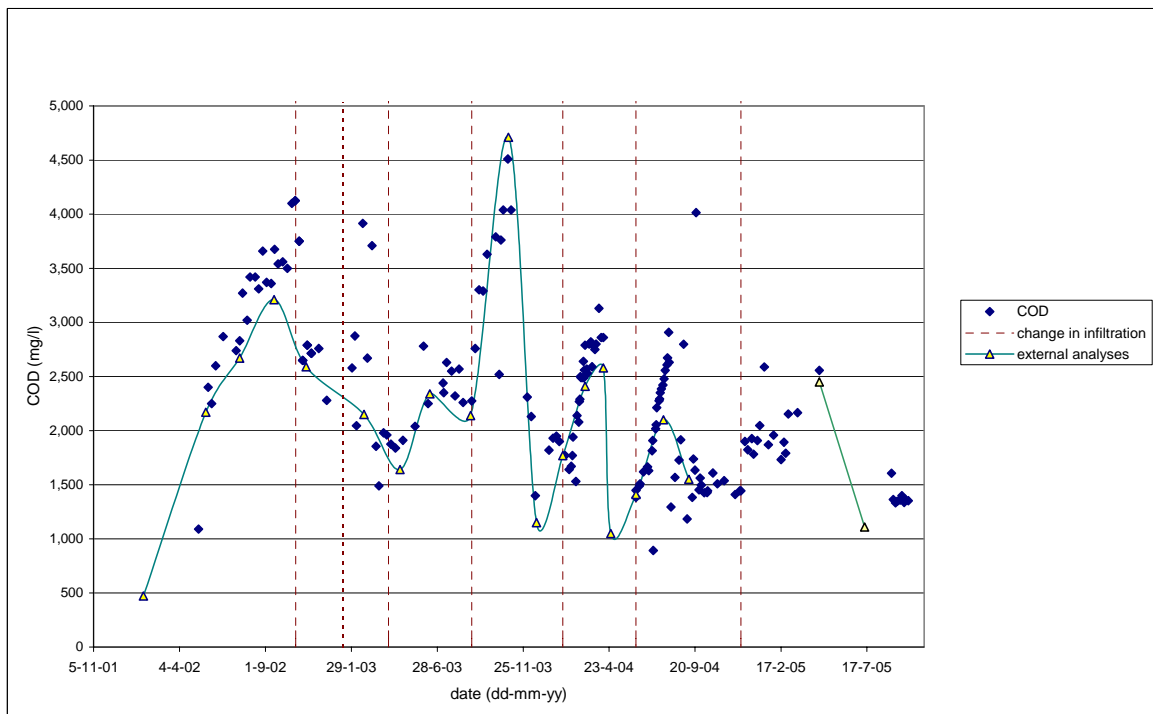
Figure 20   COD levels in the drained leachate in the period spring 2002 to spring 2005

The concentrations of N_{Kj} are presented in Figure 21. Although the picture shows similarities with Figure 20, the overall decrease in concentration appears to be absent in this case.

The overall picture of the concentrations occurring in the leachate in comparison with the criteria defined in Annex II of the Landfill Directive for inert waste (*2*) is presented in Table 4.

Additional measures for leachate management

Table 4 indicates that the standards for inert waste landfills have not yet been met for DOC, Cl$^-$ and for the metals As, Cr and Ni. Se levels are also shown to be too high.
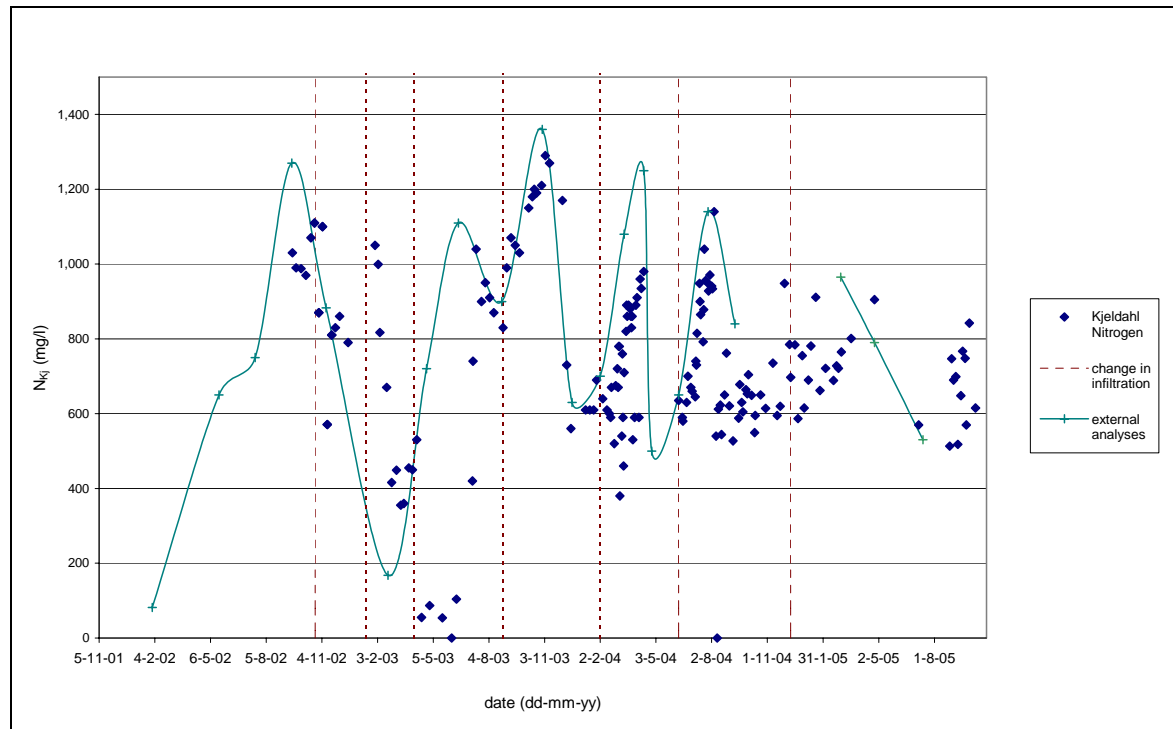
Figure 21 N_{Kj} levels in the drained leachate.

Table 4   *Comparison of Landfill Directive's Annex II criteria for inert waste with present concentrations.*

Component	Symbol	Leachate Avg. concentrations 2005 mg/l	Limit value C_0 column test mg/l	Ratio Avg.conc/ C_0
Arsenic	As	0.143	0.06	2.38
Barium	Ba	1	4	0.25
Cadmium	Cd	0.001	0.02	0.05
Chrome total	Cr	0.232	0.1	2.32
Copper	Cu	0.01	0.6	0.02
Mercury	Hg	0.0001	0.002	0.05
Molybdenum	Mo	0.014	0.2	0.07
Nickel	Ni	0.18	0.12	1.50
Lead	Pb	0.015	0.15	0.10
Antimony	Sb	0.01	0.1	0.10
Selenium	Se	0,01	0.004	2.50
Zinc	Zn	0.075	1.2	0.06
Chloride	Cl^-	1,400	460	3.04
Fluoride	F^-	-	2.5	-
Sulphate	SO_4^{2-}	479	1,500	0.32
Fenolindex		-	0.3	-
Dissolved Organic Carbon	DOC	707	160	4.42
Total dissolved solids	TDS		-	-

Beyond the criteria listed in Annex II, the values for nitrogen are also of concern. A target value for the final concentration can be set at 11.3 mg N/l (corresponding to 50 mg NO_3^-/l), either in the form of ammonium or nitrate (to ensure compliance with the Groundwater Directive). This indicates that the required reduction ratio for nitrogen would be 9,000%, which is considerably higher than for the elements listed in Table 4. To overcome this problem, an additional leachate treatment unit was incorporated into the recycle loop for the Landgraaf pilot. This unit has been in operation since July 2005. Experience so far shows that the conversion of ammonium to nitrate is achieved successfully at a conversion rate of 90%. The nitrified leachate is sent back to the landfill, where denitrification under anaerobic conditions is expected to take place. The effect of denitrification cannot yet be verified, although the nitrate levels in the leachate have remained negligible. Additional modelling has been set up to quantify the long-term effect of this treatment. Results will become available shortly. These will also incorporate the effect of flushing the pilot.

Flushing of the pilot

For the other elements mentioned in Table 4, flushing is an important measure to reduce the concentrations to the required level. For these metals it cannot necessarily be assumed that their concentrations will reduce with flushing, as their concentrations may also be determined by solubility. In addition, complexation with DOC could have a great influence on their concentrations. The correlation between the relevant metal concentrations and DOC concentrations has been investigated for the Landgraaf Pilot. The results are shown in Figure 22.
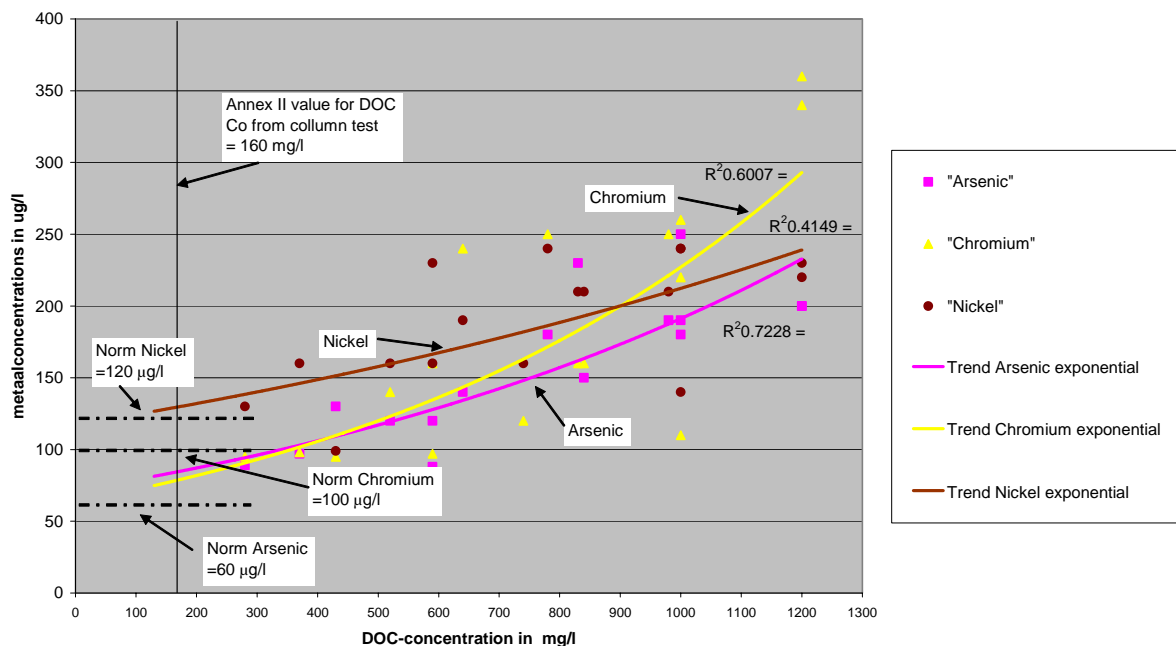

Figure 22   Metal concentrations in leachate versus DOC concentrations

The trends presented in the figure suggest that for the metals in question a correlation exists between the metal concentrations and DOC-values. If this correlation proves to be valid, the metal concentrations, with the possible exception of arsenic, could be reduced sufficiently by flushing of DOC.

Making use of the model developed to describe the biochemical and hydrological parameters, as presented in sections 3.6 and 3.8, calculations were performed to determine the expected changes in the leachate concentrations.

For these calculations the following scenario was used for the hydrological parameters:
Period 1:   0 – 5 years, recirculation flow equivalent to infiltration at 750 mm/yr, included in this is a supplement corresponding to 300 mm/yr. The amount of additional water added is also removed from the cell (discharged).
Period 2:   5 – 20 years, infiltration rate at 750 mm/yr, completely consisting of additions of clean water. This period is characterised by flushing, i.e. no recirculation.
Period 3:   20 – 30 years, infiltration rate at 300 mm/yr, also consisting of clean water, with no recirculation.

The changes in concentration for the main constituents of the leachate are presented in Figure 23.
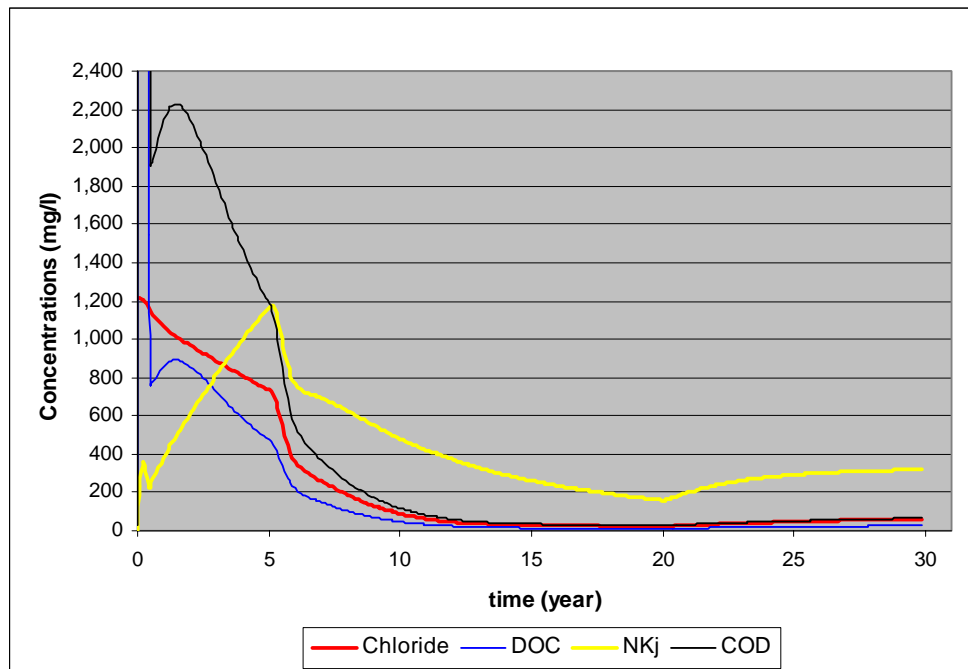
Figure 23   Changes in the concentrations of Cl^-, Kjeldahl nitrogen (N_{Kj}) and Dissolved Organic Carbon (DOC)

Two main conclusions can be drawn from Figure 23:
- By applying this method of operation, the DOC concentration can be reduced to a level where the concentrations of the heavy metals should no longer constitute a problem.
- The reduction of concentrations achieved by flushing is much less pronounced for N_{Kj} than for the other components.

In order to investigate the possibilities of a further reduction in the long-term organic nitrogen concentrations, additional model calculations were performed. The scenarios that were evaluated have the same hydrological characteristics as that mentioned above. Changes were made with regard to the possibility of biological treatment (nitrification/denitrification) of the recirculated flow and with respect to the life expectancy of the anaerobic biomass, which is

capable of temporarily storing a certain amount of nitrogen. The scenarios can be described as follows:

Scenario A   During the first 5 years the recirculation and treatment is increased to 1,500 mm/yr, the supplement (and discharge) is left at the value of 300 mm/yr. After 5 years the supplement is increased to 750 mm/yr, recirculation and treatment is set at 900 mm/yr. After 20 years, recirculation and treatment is stopped and the supplement is reduced to 300 mm/yr (natural precipitation).

Scenario B   Identical to Scenario A, except that the half-life of the anaerobic biomass has been reduced from 4 years to 1 year. This could possibly be achieved through aeration of the landfill.

The results of these two scenarios are presented in Figure 24.
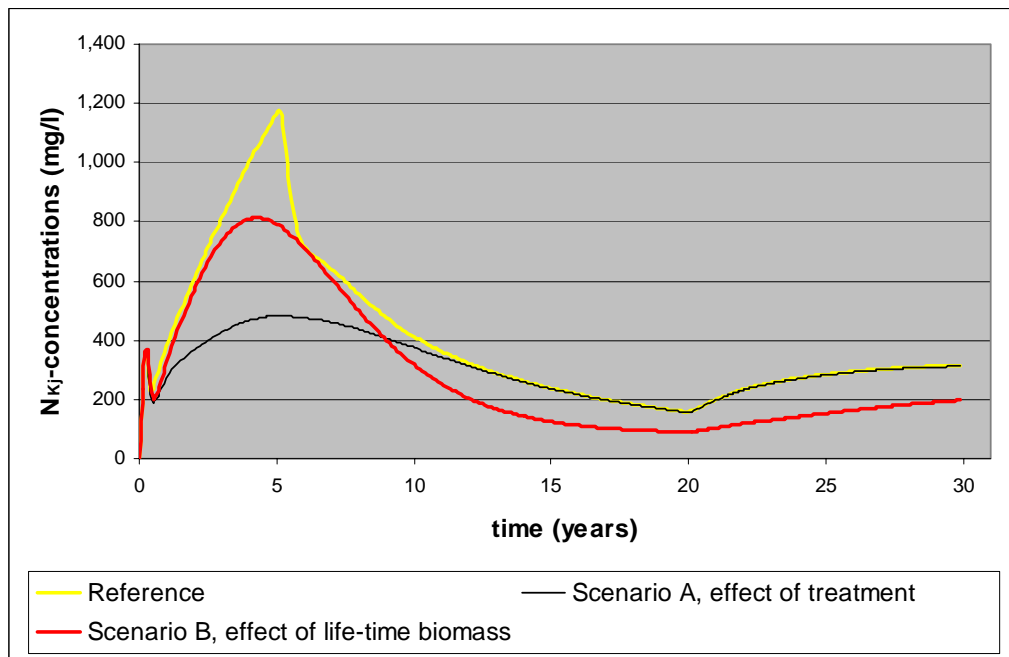
Figure 24   Effect of leachate treatment and biomass lifetime on N_{Kj} concentrations

The results presented in Figure 24 indicate that the treatment of recirculated leachate is quite effective in reducing the N_{Kj} concentrations in the operational phase of the landfill. In this way, it can contribute significantly to reducing the costs associated with the discharge of leachate. In the long run the effect of treatment reduces and the transfer of N_{Kj} from stagnant zones and from decaying biomass becomes dominant in the same way as for the reference case. The indirect effects of the introduction of nitrified leachate into the cell have (not yet) been incorporated into the model, but these are expected to create an additional reduction in N_{Kj} concentrations.

Reducing the lifetime of the biomass is an important step in achieving a more favourable course for the N_{Kj} concentrations in the long-term. If this is achieved by aeration, part of the nitrogen may be removed from the cell by a stripping effect. This possibility has been demonstrated by Ritzkowski (*24*), in tests in simulated landfill reactors. In these tests a significant reduction of N_{Kj}-concentrations was observed. It was determined that 50% of this reduction resulted from the stripping of ammonia from the leachate.

The application of these mechanisms will be the subject of further research in the Landgraaf Pilot.

Landfill gas management

The landfill gas quality during the first year was quite good (50% methane). It deteriorated during the last year to low levels of methane (20% methane). The calculated gas production (projected) seems to be an overestimation, probably caused by an overestimation of the biodegradable part of the organic waste, an aerobic composting phase during preparation of the waste (size reduction and homogenisation by the compactor), mixing (partly outside the bioreactor) of the waste and the long period of time taken for filling in relatively thin layers. The landfill gas production curve is presented in Figure 25.
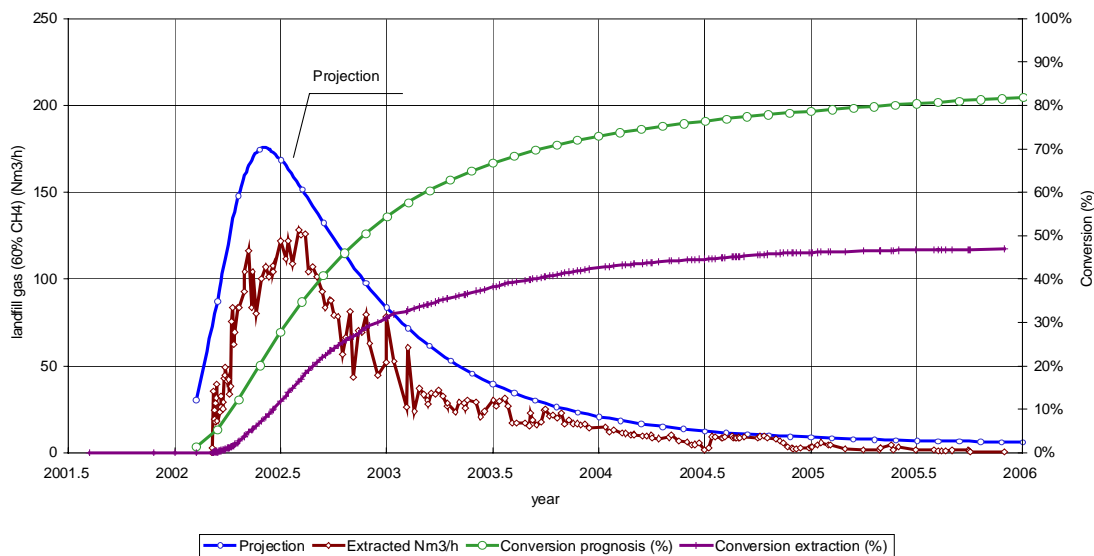
Figure 25   Landfill gas production ('adapted': recalculated to 60% of methane)

Methane emissions and methane oxidation were calculated and measured. The emission was measured as 200 dm^3 CH_4/h, or approximately 2% of the extraction rate at the time. This emission is quite low and was related to one small area (the actual oxidation efficiency could only be measured in this area.) Over the rest of the surface no methane concentrations were found in the top cover and hence no oxidation rates or efficiencies could be determined. The average oxidation rate was estimated to be 0.5 dm^3 CH_4/m^2h. The oxidation efficiency was calculated to be between 35 and 60%, measured by the ^{13}C method, for the points with measurable methane concentrations.

Research cited previously ((*25-27*) and others) indicates that for a standard top cover with vegetation methane oxidation rates between 0.34 and 5.6 dm^3 CH_4/m^2h are feasible. In a recent article (*28*) it was suggested that a surface load of 0.5 to 1.0 dm^3 CH_4/m^2h could serve as a limit for requiring active gas extraction. The present gas production at the Landgraaf Pilot is estimated to be 3.7 m^3 STP CH_4 /h (Standard Temperature and Pressure, 273.15 K and 101,325 Pa), equivalent to a surface load of 0.93 dm^3 CH_4/m^2h. This indicates that after only four years of operation, gas production is reduced to a level that can be dealt with by oxidation in the top cover.

The difference between the projection and the actual extraction was smaller than approximately 20 m^3/h, after the first half year of operation of the gas extraction. The resulting load on the oxidation layer would have been 3 dm^3 CH$_4$/m^{2}h. Given the low level of measured emissions in May 2003, the actual oxidation rate may have been in this order of magnitude. In the first half year of operation the potential load on the oxidation layer may have been higher, as the extraction system had not been fully established.

This first part of the curve has the highest uncertainties with respect to possible methane emissions. In order to be sure that no such emissions occur, operation of an air injection system during the build up of a landfill cell could be considered, which would prevent the landfill from becoming anaerobic before proper gas extraction can take place. In the case of the Landgraaf Pilot, passive emissions from the vertical gas extraction wells indicated there being only very small concentrations of methane (<1% CH$_4$).

More information about the research on organic processes is presented in the research report Bioreactor (*29*), which is also referenced in section 6.3.

4.3   Equifill: predominantly inorganic

Remarks on achieved L/S ratio in pilot

At the end of the project, the calculated L/S in the Nauerna Pilot turned out to be too high. The reason for this deviation was that air could enter the flow meter, thereby causing too high water volumes to be measured. This problem has now been solved and the magnitude of the error will be addressed in order to correct the L/S ratio in later stages. The reported L/S ratios in this report are therefore an overestimation of the real values.

The effect of correction of the L/S ratio on the calculated emissions is shown in Figure 26. The blue diamonds show a result of a laboratory column leaching test. In this example the L/S ratio was lowered by a factor of 10. It can be seen that the data mainly shifts along the original leaching curve. The effect on the calculated emission is limited to a factor of approximately 2. This implies that the interpretation of the results will not change when the L/S value is corrected at a later stage. The comparison of emissions with the Landfill Directive's criteria are based on the results of laboratory leaching tests at L/S=10 and not on the measurements from the pilot. Therefore, the conclusions from the comparison are also unaffected.

Leachate quality

The leachate quality from the pilot and the lysimeters, in comparison with the testing at laboratory scale (see Figure 27, all other elements are described in the Dutch report research report 'Equifill'), shows that for the waste mix studied the quality objectives for inert waste, as laid down in Annex II of the Landfill Directive, were met with the exception of Cl$^-$ and SO$_4^{2-}$ (see Table 5). In general, the emissions measured at laboratory, lysimeter and pilot scale agree well with each other. Significant differences were found for Cu, Fe, Mn and SO$_4^{2-}$. These elements will be specifically addressed in the section entitled 'Influence of redox potential on the emission of contaminants.' It should be noted that the observed differences between the laboratory and field measurements do not influence the interpretation of the regulatory aspects.
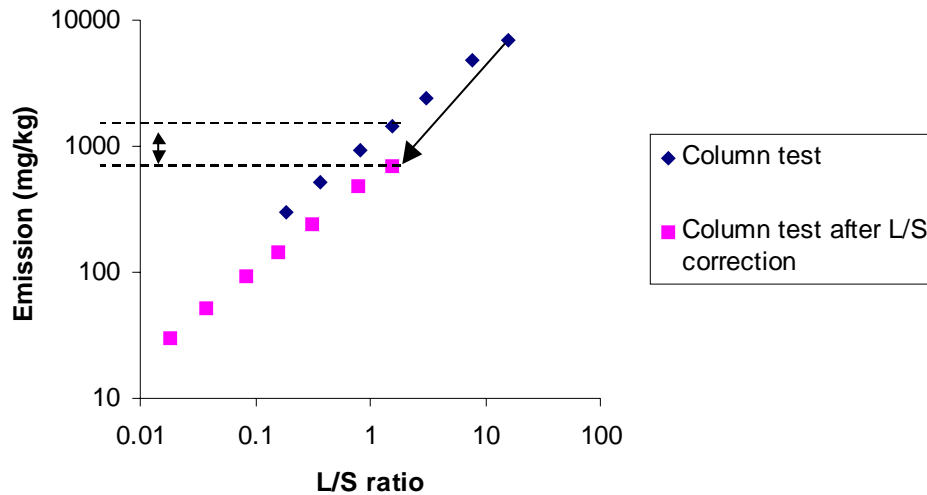
Figure 26   Influence of L/S ratio correction on the calculated emission of contaminants. In
             this example the L/S ratio of the laboratory column leaching test (blue diamonds)
             was corrected by a factor of 10 (pink squares).
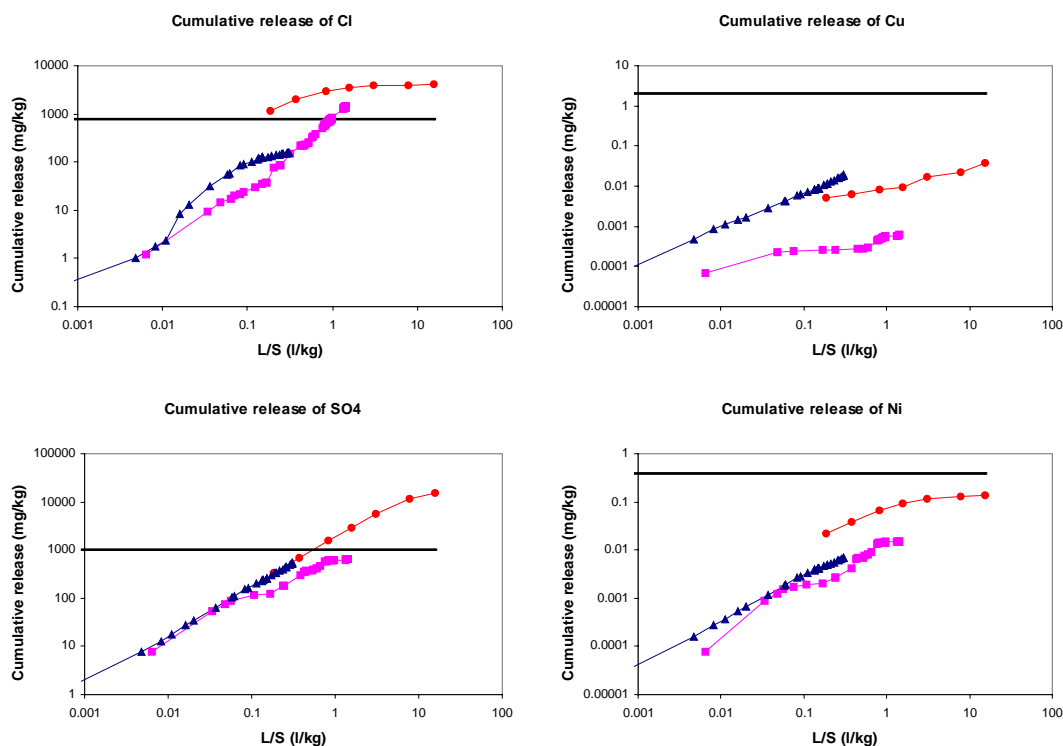
Figure 27   Cumulative leaching of Cl⁻ , Cu, SO₄²⁻and Ni as a function of L/S for experiments
             on laboratory (circles), lysimeter (triangles) and pilot scales (squares). The
             horizontal line represents Annex II's waste acceptance limit for inert waste.

Given that there are currently no specified criteria for organic micro chemical pollutants, these
could not be properly assessed. Leaching of organic micro chemical contaminants can be
handled and assessed in much the same way as inorganic contaminants (*30*). For poorly-

water soluble organic contaminants, release is largely determined by (sub-fractions of) DOC as can also been seen in Figure 28.

Current modelling capabilities allow behaviour to be verified prior to a new waste being accepted in order to influence leachate quality. It is possible to model the effects of increased element loading in the input e.g. Pb (*23*). The chemical speciation occurring provides detailed insights into the chemical forms controlling release in the pH domain relevant for the waste mix.
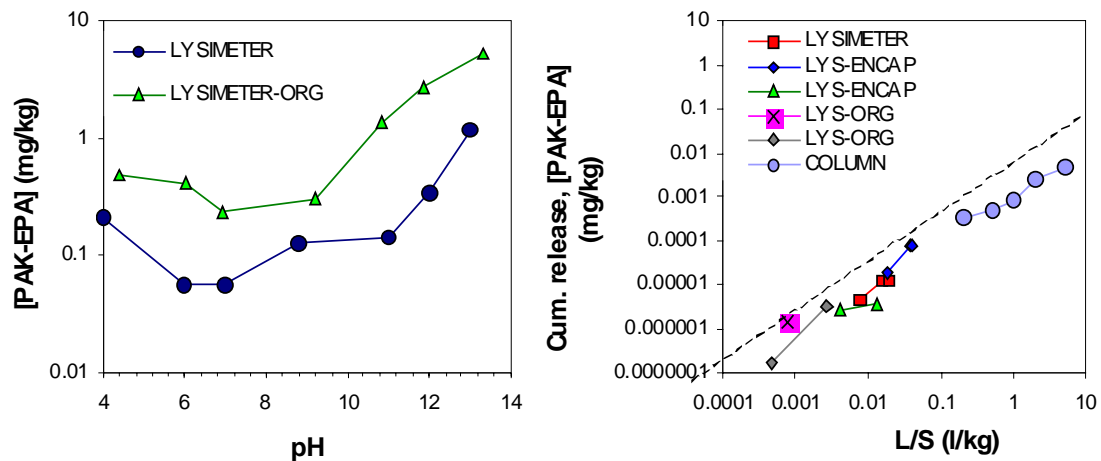
Figure 28   Leaching of organic micro chemical pollutants as a function of pH (pH-static leaching test) in the predominantly inorganic waste mixture (lysimeter) and the waste mixture enriched with 10% organic waste. The right graph shows the leaching as a function of L/S at different scales of testing (lysimeter and column leaching test).

Regulatory aspects

In Table 5, the emissions from predominantly inorganic waste are compared to the acceptance criteria for inert waste. It can be seen that the criteria were met for most elements with the exception of Cl^- and SO_4^{2-}. When an L/S of approximately 1 to 2 is reached after 30 years, a significant portion of the leachable Cl^- will have been washed out. The results show Sb leaching to be critical compared to the acceptance criteria for inert waste. However, measured concentrations were generally at or below the limit of detection (about 5 ppb with ICP-AES). This implies that additional measurements with hydride generation and atomic fluorescence spectrometry (HAFS) should be carried out to assess whether Sb is a critical parameter. Hence, on the basis of these results, we cannot properly conclude whether Sb will be a critical element with regard to the acceptance criteria for inert waste landfills. However, the reported emissions analysed by ICP-AES were within the upper limits for the Sb emission.

This implies that SO_4^{2-} may be the only remaining critical component. Its concentration is related to gypsum solubility control, which cannot be changed. The solubility-controlled leaching of SO_4^{2-} will continue until all the gypsum present in the landfill is dissolved. The gypsum content can be lowered, but it is not possible to eliminate this compound from the incoming waste stream. Solubility control by gypsum extends well beyond the L/S ratio of 1 to 2 projected to be reached at closure. Flushing during the operational phase will not sufficiently reduce the SO_4^{2-} concentrations in the leachate. This aspect is common to all landfills. It would seem that dilution in the groundwater remains the only subsequent control measure.

Table 5   *Comparison of emissions from predominantly inorganic waste (based on laboratory column leaching test) with the waste acceptance criteria for inert waste.*

Element	Waste mix L/S=10 mg/kg	Inert waste acc.crit. L/S=10 mg/kg	Ratio	Remark
As	0.099	0.5	0.20	
Ba	0.534	10	0.05	
Cd	0.031	0.04	0.76	
Cl⁻	4,029	800	5.04	
Cr	0.023	0.5	0.05	
Cu	0.038	2	0.02	
DOC	353	500	0.71	
Mo	0.123	0.5	0.25	
Ni	0.136	0.4	0.34	
Pb	0.323	0.5	0.65	
SO_4^{2-} as S	5,486	333	16.5	
Sb	0.064	0.06	1.07	Concentrations at DTL
Se	0.053	0.1	0.53	Concentrations at DTL
Zn	0.833	4	0.21	
Hg	0.0022	0.01	0.22	Extrapolation from pilot data
F	7.9	10	0.79	Extrapolation from pilot data

Influence of redox potential on the emission of contaminants

Long-term changes in leaching behaviour beyond what has been studied here are unlikely as the combination of test conditions covers all conditions that can be expected. The monitoring of the leachate and runoff quality in the pilot has been underway for over four years. It has been shown (*12*) that in general there is good agreement between the leaching behaviour of the constituted waste mixture and the leachate obtained from the full scale pilot. This indicates that in spite of an apparently very heterogeneous mix of materials, the leaching of many constituents is determined by well defined solubility controls. However, widely different redox potentials were measured in the laboratory, lysimeters and pilot (range roughly from -300 mV to +300 mV), as can be seen in Figure 29. The leachate from the pilot has had a relatively constant redox potential of about -200 mV, whereas the lysimeter experiments started with a redox potential within the same range. About 1.5 years after the start of this experiment, the redox potential in the lysimeter increased to more or less stable values of approximately +200 mV. Presumably, the waste material was oxidised by direct exposure to the atmosphere. The laboratory column leaching tests exhibited an increasing redox potential ranging from -100 mV at the start of the experiment, to values of about +250 mV at the end (after 16 days). In the latter case the leachant was oxygen-saturated water, which implies that the reducing capacity of the waste mix was limited. The effects of different redox conditions on the leaching behaviour of several relevant elements are discussed below.
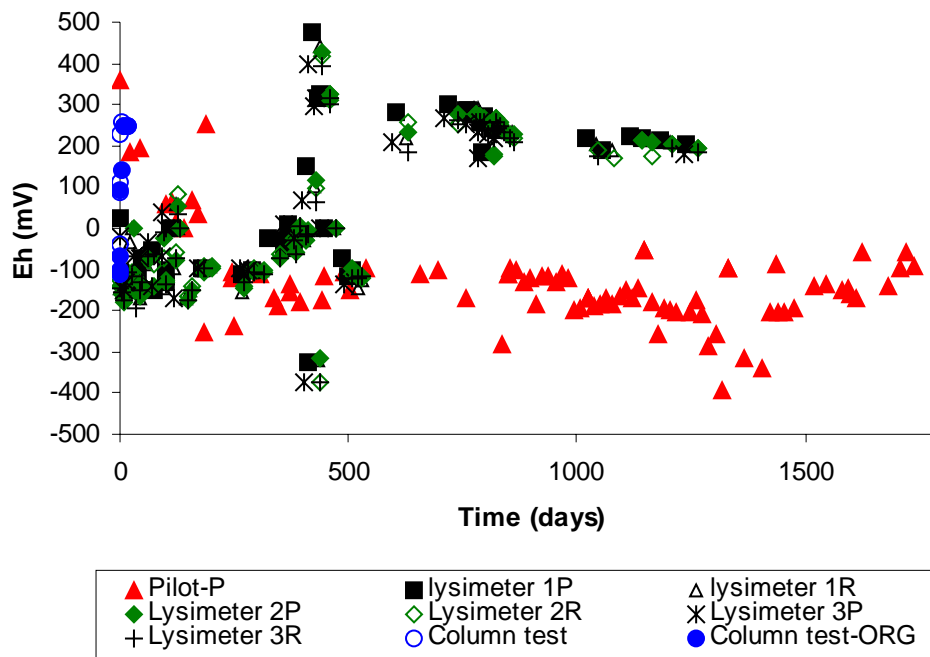
Figure 29   Redox potential measurements (uncorrected measurements) as a function of time
in laboratory (column test), lysimeter and pilot scale experiments with
predominantly inorganic waste (*11*).

The observed differences in redox potentials in the performed experiments led to differences
in the release of elements such as Cu, Fe and Mn (Figure 30). This is still consistent with the
data presented previously from testing on different scales (*12*). The leaching of SO_4^{2-} is
presented in Figure 30 by way of example. It can be seen that the SO_4^{2-} data obtained from
the different testing scales show a very consistent pattern and a close match to the cumulative
release. The cumulative release of Cu was much lower in the pilot compared to the other test
types (lysimeter and column leaching tests). Fe and Mn showed relatively high leaching in the
pilots, this release is in line with the laboratory column test results but was much higher than
those obtained in the lysimeter experiments. The reason for these differences is probably due
to the difference in redox potentials between these leaching tests. In general, the redox
potential in the leachate from the pilot was around -100 to -200 mV. Given that the leachate
collection system is not sealed, the redox potential in the pilot cell may actually be lower. The
redox potential in the lysimeter experiments was 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 lower for Cu(I) species than Cu(II). Moreover, the formation of relatively
insoluble Cu 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, resulting in an enhanced leaching of the
total Fe. The column test results also showed a relatively high Fe leaching. This might also
have been due to a reducing environment in several fractions of the column. The extent of
reduction during a leaching test depends on the reducing properties of the material, which can
be measured with a Cerium titration method (NVN 7348).

The leaching of Mn in the pilots and the column experiments was higher than the in the lysimeters. The soluble Mn(II) species will be dominant under reducing conditions like those in the pilot and (to a lesser extent in) the column test. This species can easily be oxidised to Mn(III) which forms solid oxides (*31*).

The leaching of Fe and Mn in the column test and the pilot were consistent. The leaching of these elements in the lysimeter experiments was significantly lower. However, Cu leaching in the column test was consistent with the lysimeter experiments. The Cu leaching in the pilot was 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 was generally relatively high in comparison with the standard redox potentials of the Cu, Fe and Mn reduction reactions. The eluates collected in the column test had 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 were presumably low enough to reduce Cu also. Preliminary calculations using predominance diagrams also indicate that Cu(I) species are likely to be present under these conditions.
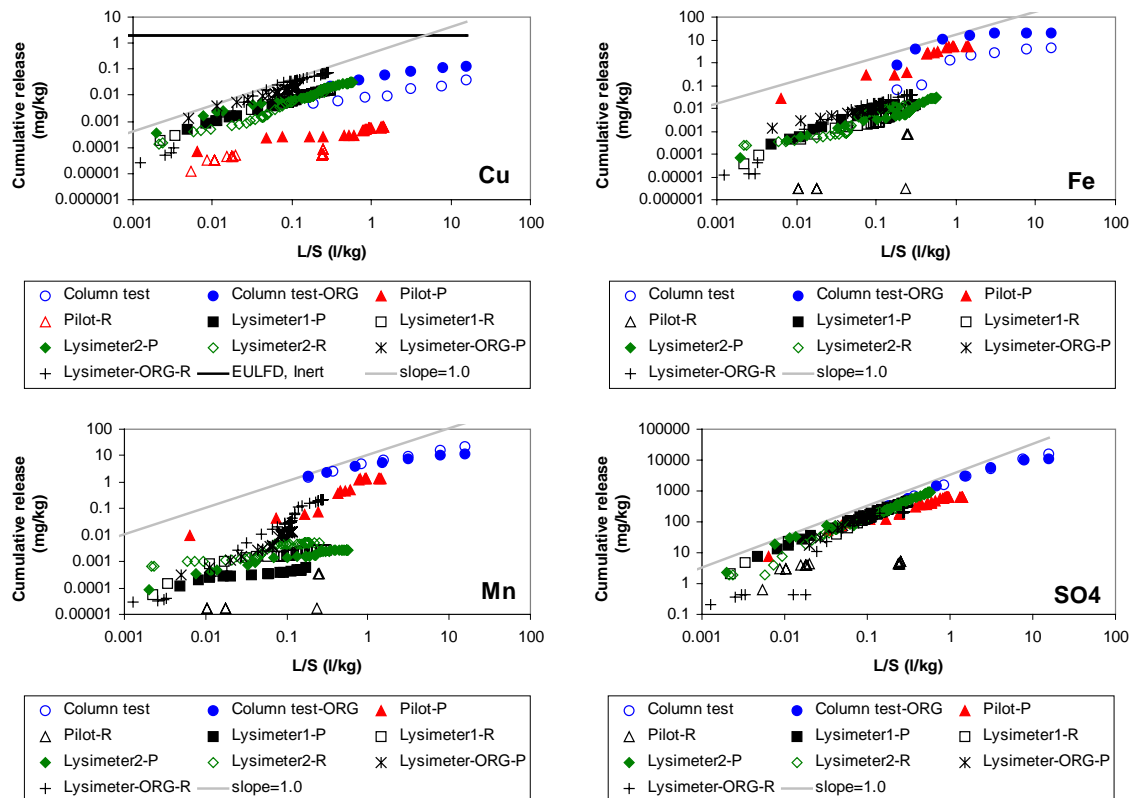
Figure 30   Cumulative release data as a function of L/S from column leaching tests (Column test and Column test-ORG), lysimeter experiments (Lysimeter1-3) and a pilot. The line with slope = 1 shows the slope of a solubility-controlled release and the black line for Cu indicates the Landfill Directive's limit for inert waste.

The noticeable differences are for NH_4^+, which was completely oxidised, and Mn and Fe, which were both low compared to the full scale pilot. The release of Cu increased due to the oxidising conditions in the lysimeters, but the emissions are still 1 to1.5 orders of magnitude lower than the Annex II criteria.

Model parameters

The availability of all the elements used as inputs for geochemical speciation modelling is given in Table 6. It should be noted that availability was determined as the maximum concentration obtained in the pH-static leaching test. The availability of Al and Pb was changed in the initial speciation calculations. The leached concentrations as measured in the pH-dependent test (lowest pH around 4) were not sufficient to properly account for their availability.

Table 6   *Available concentrations of elements used in geochemical modelling. Additional parameters to describe binding to the solid phase were: HFO (4.3E-3 kg/kg), total humic acid content (1.03E-3 kg/L) and clay (0 kg/kg) (23).*

Element	Availability (mg/kg)	Element	Availability (mg/kg)	Element	Availability (mg/kg)	Element	Availability (mg/kg)
Al	3,000.00	Mg	3,002.13	CO_3^{2-}	55,000.00	SO_4^{2-}	12,715.33
As	2.57	Mn	573.68	Cr	19.19	Sb	0.39
B	18.65	Mo	2.87	Cu	39.77	Se	0.32
Ba	7.54	Na	1,400.00	F^-	50.00	Si	3,014.79
Br	34.52	NH_4^+	609.57	Fe	16,360.59	Sr	176.10
Ca	50,151.07	Ni	23.23	K	1,158.57	V	5.22
Cd	2.76	PO_4^{3-}	81.57	Li	3.02	Zn	2,400.83
Cl^-	5,267.82	Pb	251.00				

DOC plays an important role in the mobilisation of various contaminants. However, DOC is a sum parameter of all the organic carbon species. The NICA-Donnan model (*16*) was used to account for the complexation of contaminants to solid and dissolved humic acid. It was assumed that 20% of DOC consists of humic acid, over the whole pH range. ORCHESTRA calculates the geochemical speciation from pH 1 to 13 with intervals of 0.2 pH values. The DOC data was fitted to a polynomial function in order to describe the pH-dependent leaching of DOC. The calculated DOC concentrations at pH intervals of 0.5 are given in Table 7.

Table 7   *Calculated DOC concentrations as a function of pH, based on measurement in pH-static leaching tests (23)*

pH	DOC (kg/l)	pH	DOC (kg/l)	pH	DOC (kg/l)
1	1.96E-05	5.5	1.06E-06	10	7.47E-06
1.5	1.75E-05	6	1.62E-06	10.5	1.01E-05
2	1.44E-05	6.5	2.44E-06	11	1.5E-05
2.5	1.09E-05	7	3.31E-06	11.5	2.32E-05
3	7.54E-06	7.5	4.1E-06	12	3.63E-05
3.5	4.76E-06	8	4.71E-06	12.5	5.6E-05
4	2.71E-06	8.5	5.15E-06	13	8.44E-05
4.5	1.46E-06	9	5.55E-06	13.5	0.000124
5	9.49E-07	9.5	6.17E-06	14	0.000178

Table 8   *Possible solubility controlling minerals selected from initial speciation calculations.*
             These minerals were subsequently used as inputs for the model predictions (23).

Mineral name	Mineral name	Mineral name	Mineral name
Albite[low]	Gypsum	Ferrihydrite	Otavite
AlOHSO$_4$	OCP	Brucite	Hydromagnesite
Boehmite	alpha-TCP	MnHPO$_4$[c]	Strontianite
Leucite	Ca$_2$Cd[PO$_4$]$_2$	PbMoO$_4$[c]	Cr[OH]$_3$[a]
BaSrSO$_4$[50%Ba]	Cd[OH]$_2$[c]	Bunsenite	Manganite
Ba[SCr]O$_4$[96%SO$_4$]	PbCrO$_4$	Pb$_2$V$_2$O$_7$	Ba$_3$[AsO$_4$]$_2$
Anhydrite	Cu[OH]$_2$	Willemite	Sb[OH]$_3$
CSH_ECN	Fluorite	Zincite	
Ca$_2$Pb[PO$_4$]$_2$	FCO$_3$Apatite	Calcite	

Initial speciation calculations indicated several possible solubility controlling minerals. The selected set of minerals used for the model prediction calculations are shown in Table 8. In some cases more than one mineral was selected, based on possible solubility control in different pH ranges.

Comparison of model with experimental results

The geochemical model results for all the elements considered are compared with the results of the pH-static leaching test in Figure 31 to Figure 33. In general, the model describes the leaching behaviour of the waste mixture quite well; especially given the fact that changes to the input parameters may have affected the predicted behaviour of several other elements. This implies that the freedom available to vary input parameters is limited dramatically by taking all elements into account simultaneously. Given that the model assumes equilibrium, and it is known that equilibrium is not reached within 48 hours contact time, the kinetics of dissolution and precipitation was a factor to be considered when assessing the results. These effects resulted in an apparent deviation from the model prediction, as the leached concentrations may have increased or decreased due to equilibrium not having been reached and/or kinetic processes. Recent work (*32*) has shown these effects in relation to the material's own pH, where the system is closest to equilibrium. Therefore, it is assumed that there is sufficient understanding of the chemical processes that determine the leaching behaviour in this waste mixture.

The pH-static leaching data and geochemical model results for Al, Ba, Ca, Fe, Mg, Si, SO$_4^{2-}$ and Sr are presented in Figure 31. There are significant deviations in the model description compared to the actual leaching data, such as the deviation of more than one order of magnitude for Si at pH>11. Apparently, the chemistry in this pH range is not yet fully understood. The model underestimates the leaching of Fe by one to two orders of magnitude in the pH range 5 to 7. The deviations for Mg, Sr, SO$_4^{2-}$ and Ba are expected to be largely related to kinetics.

Despite the deviations from the model for the leaching of the described elements, the overall prediction of the chemistry of the major elements is an important finding and provides a good basis for the understanding of the chemical processes occurring in these apparently very heterogeneous materials.

Figure 32 shows the pH-dependent leaching behaviour and results from geochemical modelling for Cd, Cu, Mn, Ni, Pb, Zn, Cl⁻, K and Na. It can be seen that Cd leaching is

generally overestimated over the whole pH range by up to one order of magnitude. Although Cu leaching is described adequately at both low and high pH, there is a relatively large overestimation in the pH range from 5 to 9. Leaching of Cu is predominantly controlled by complexation to humic and fulvic acids in this pH range. However, the modelling results are based on the assumption that 20% of DOC consists of humic acid over the whole pH range. Earlier measurements of humic and fulvic acid concentrations as a function of pH in the leachate from a compost sample, have shown that the leaching of humic acids in particular is strongly pH-dependent (unpublished results, ECN). This may explain the general overestimation (pH range 5 to 9) of the concentrations of the heavy metals that have a high affinity for complexation to humic and fulvic acids (Cu, Ni, Cd and Pb). The leaching behaviour of Mn and Zn are reasonably well described. Pb is well described in the neutral pH range, but dissolution kinetics may be the cause of the discrepancy between the model and the measured result in the mildly acidic and mildly alkaline ranges. The description for Ni is not yet adequate in the pH range 4 to 9, apparently a relevant mineral phase or chemical process is missing in the model.

The pH-static leaching data and geochemical model results for As, B, CO_3^{2-}, Cr, Mo, Sb, PO_4^{3-}, Se and V are presented in Figure 33. The degree to which the model describes the leaching data of these oxyanions is variable. The leaching behaviour of Cr is quite well described, whereas both Mo and V are only reasonably well described. The latter are both controlled by Pb and thus any change in Pb partitioning will affect Mo and V leachability. For B, Sb and Se there are very limited thermodynamic data, which hampers the ability to produce a proper description of these elements with this type of modelling. In the case of As, a mineral phase assumed to be present at high pH is apparently not formed in the system, thereby leading to an underestimation of its concentration. Sorption to Fe-oxide is seen as would be expected for As. The predicted occurrence of carbonation is overestimated at low pH, but can be explained by CO_2 not being allowed to escape. The underestimation at a mildly alkaline pH may require some further work. Additional work also needs to be undertaken for phosphate. The thermodynamic database is adequately stocked, but the selection of relevant mineral or sorptive phases to be included in the predictive modelling does require further work.

The partitioning between the dissolved and particulate phases, as obtained from the modelling carried out in this work provides a significantly increased insight into the mutual relationships between the elements. This allows the prediction of release behaviour under conditions other than those in the laboratory.
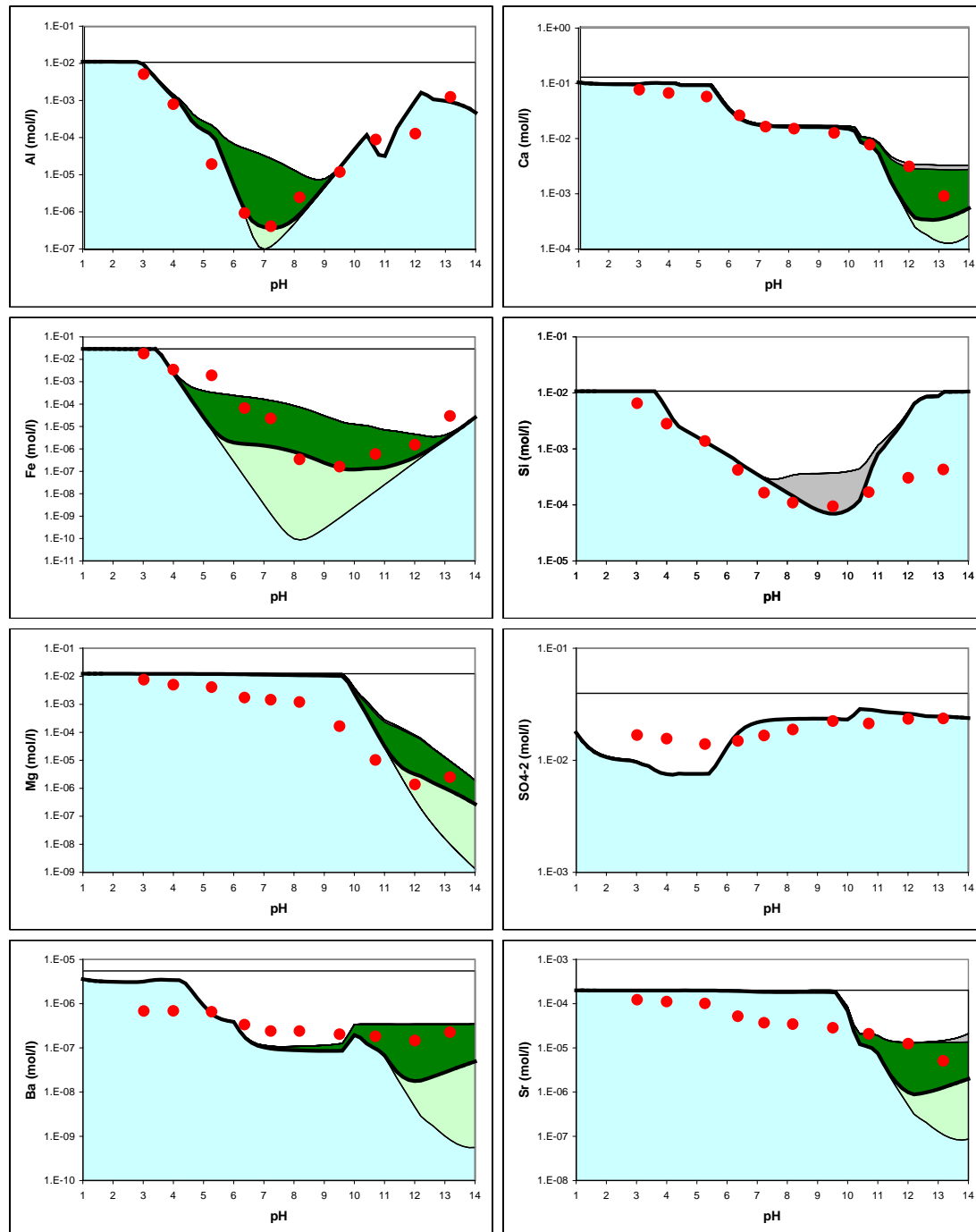
Figure 31   pH-static results and geochemical modelling for the major elements. The thick
solid line indicates the predicted concentrations in solution. The areas below this
line indicate the element speciation in solution (light blue are the free and
inorganically complexed species and light green is the organically complexed
species). Areas above the thick solid line indicate the element speciation in the
solid matrix (dark green indicates binding to solid organic matter, grey is the
amount adsorbed to hydrous ferric oxide (HFO) and the white area represents the
amount present in minerals (*23*).
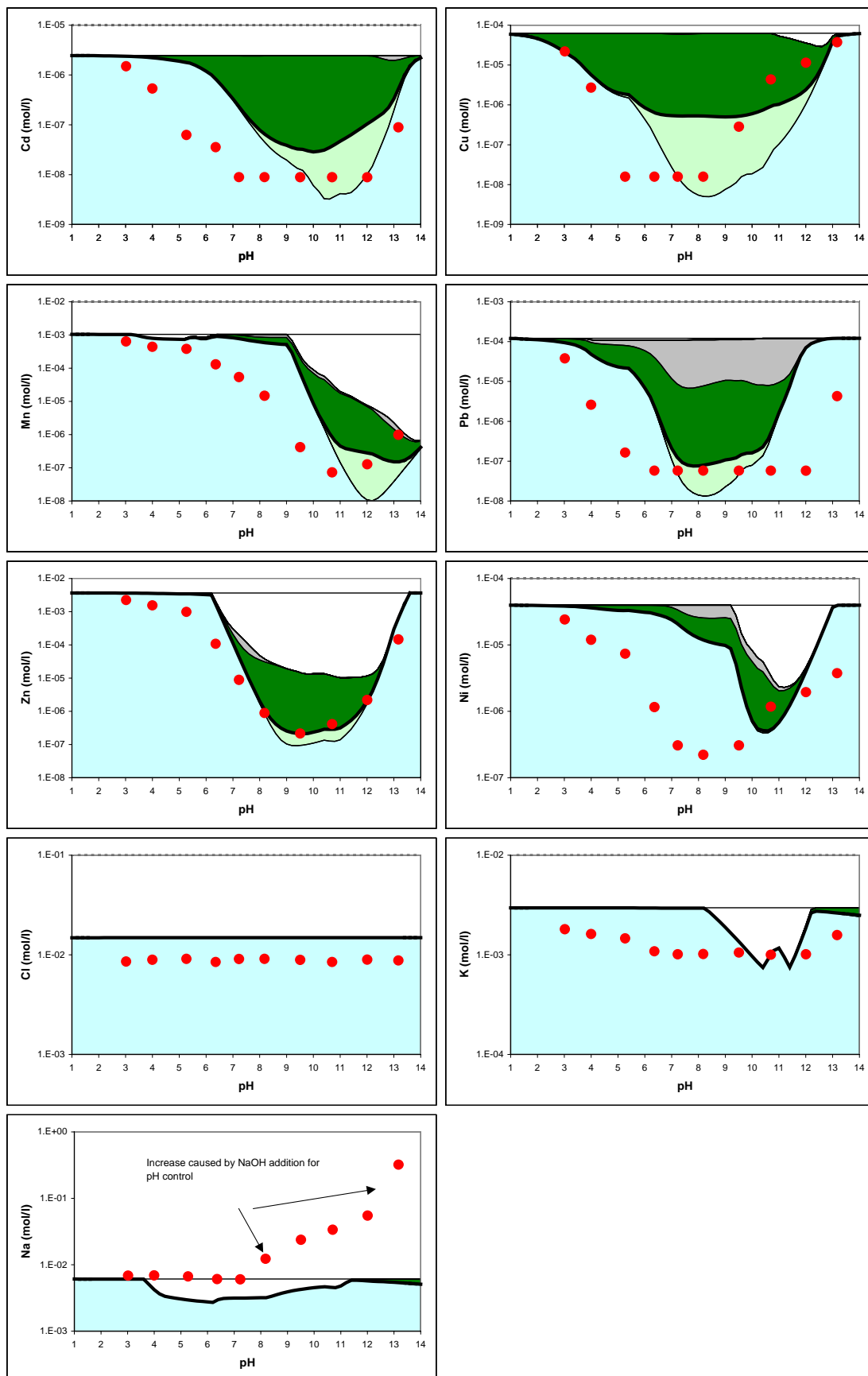
Figure 32   pH-static results and geochemical modelling for heavy metals and salts (*23*).
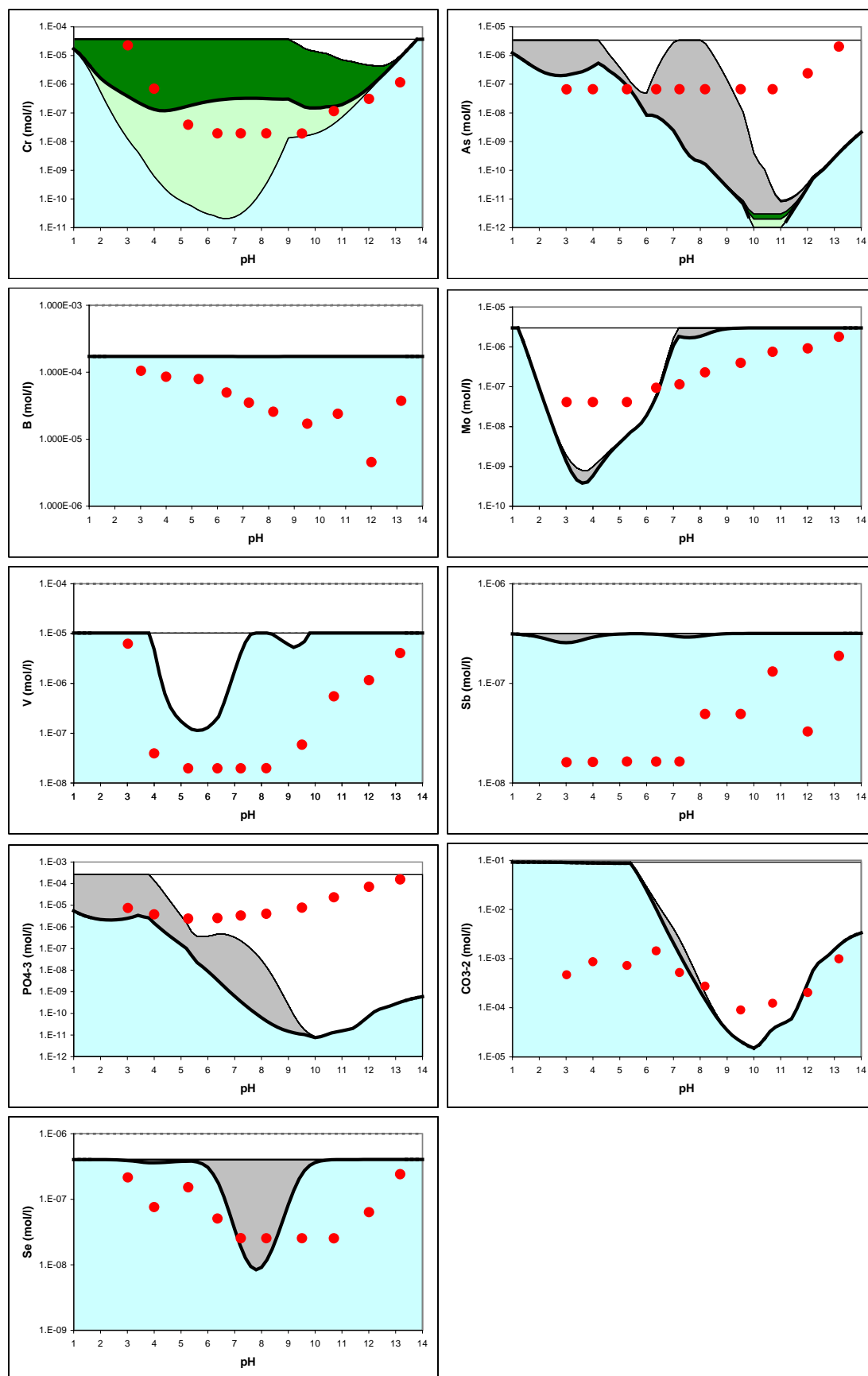
Figure 33   pH-static results and geochemical modelling for oxyanions (*23*)

Prediction of emissions in column leaching test

The release by percolation can be modelled based on the geochemical modelling of the waste mix, using the applied infiltration rate. A measure of DOC (which is still difficult to predict), is also used and is based on a similar release as K from mixed waste. Additional work is needed in order to fully understand the mechanism determining DOC leaching, and more specifically the leaching of humic and fulvic acids.

The results for Cu, Pb, Cl⁻, SO_4^{2-}, Zn and K are shown in Figure 34. The prediction is compared with the laboratory data and is shown to match rather well. This allows assessments of long-term release behaviour to be made.
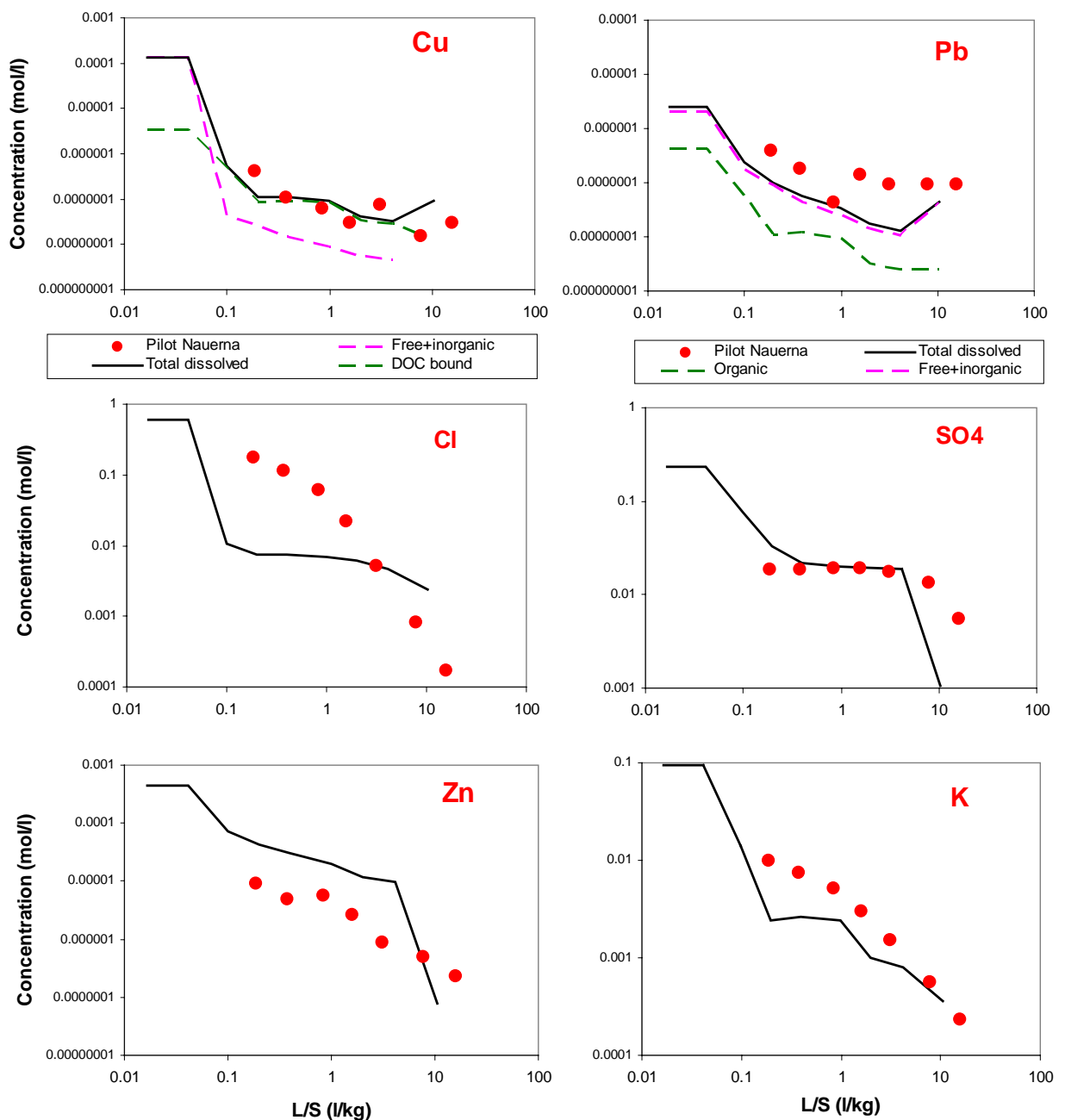
Figure 34   Measured and predicted leaching of Cu, Pb, Cl⁻, SO_4^{2-}, Zn and K in the column leaching test. The model assumes dual porosity in contrast to the organic biodegradation model as described in paragraph 3.8.

Production of landfill gas in Equifill pilot

Methane emissions were analysed by so-called 'box measurements'. Methane was collected by placing a box (0.7 x 0.8 x 0.5m) over parts of the landfill. The emitted methane was analysed by a tuneable diode laser. The methane flux was calculated to be 0.008 dm^3 CH_4/m^2h. These emissions were very low compared to e.g. a municipal solid waste landfill where fluxes of 0.15 to 3.5 dm^3 CH_4/m^2h were measured (*33,34*). In Finland and France guidance has been given by the regulatory authorities stating that a release of less than 1 dm^3 CH_4 /m^2h (or less than 10 m^3 CH_4 /ha.h) requires no further action; placement of only an oxidising cover layer may suffice.

The very low methane emissions observed in the sustainable landfill pilot for predominantly inorganic waste are consistent with the nature of this landfill concept.

More information on the research related to inorganic processes is presented in the research report Equifill (*20*), which is also referenced in section 6.3.

4.4   Monolith: immobilisation

Leachate quality

The measured pH values in both the percolate and the runoff were generally neutral to slightly alkaline, ranging from 6.8 to 9.3 (average 7.9). This is significant as the alkaline waste material has a natural pH > 12. The pH of the percolate water from the (non-pilot) stabilised waste landfill at VBM is also still neutral. This landfill has been in operation since 1998. Also, other field experiments have shown a high leachate pH of about 13 (*5-7*). Presumably the percolate and runoff only came into contact with the outer (carbonated) surface of the waste material. This could explain the relatively low pH values in the water. Moreover, the pH in the percolate water was buffered by the presence of the soil layer at the bottom of the waste compartment (*35*). Gradient relaxation in the exposed waste layer, as a result of a decreased water volume after completion of the top cover, may cause an increased influence of alkaline compounds on the percolate water quality (e.g. an increasing pH). Future work should determine whether the combination of an increased influence of alkaline compounds and a decreased water volume affects the calculated effective period of the buffering soil layer after construction of a top cover. The effect of pore sealing as a result of carbonation should also be taken into account. Fitch and Cheeseman (*6*) have identified $CaCO_3$ minerals in environmentally exposed stabilised waste. Pore sealing can lead to a lower release of alkaline components and contaminants from the waste.

The EC values in the percolate water samples (63 to 217 mS/cm) were approximately a factor of 10 to 30 times higher than the EC values in runoff samples (0.9 to 6.8 mS/cm). This can be attributed to the difference in contact times (relatively short for runoff) and possibly the depletion of the surface layer of soluble salts. Measurements on sliced core samples from the pilot indicated that at least the upper 10 centimetres had a depleted Cl^- content after 4 months.
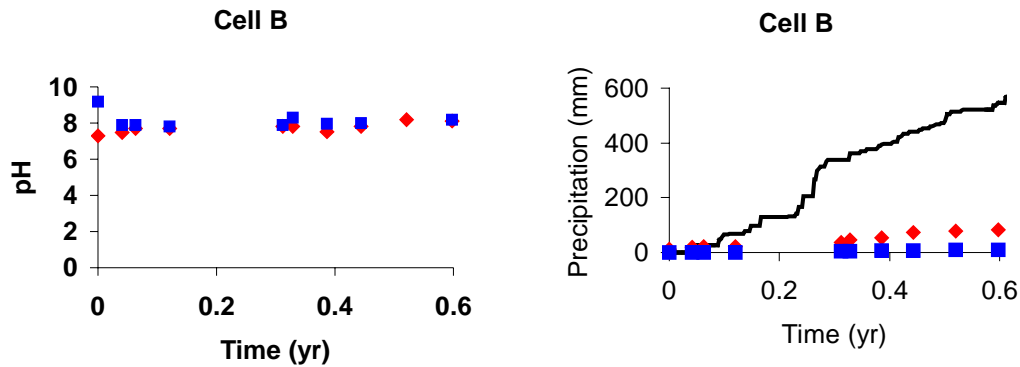
Figure 35   pH and volumes of percolate, runoff and the total precipitation as a function of time in the stabilised waste pilot. Red diamond data points represent measurements in percolate water and blue squares are measurements in runoff. The solid line is the total cumulative precipitation over time.

Release of contaminants under field conditions

The leached concentrations of Cd, Cu, Mo and Zn in the percolate and runoff are shown in Figure 36. It can be seen that the leached concentrations in the percolate water were generally higher than the concentrations in runoff, except for Zn. There were no obvious differences in the concentrations between the different compartments, which differ in height (cell A is one 1 m high, cell B and D are 3 m high) and the stabilised waste recipe used (cell A and B both contain a MSWI fly ash recipe, cell D contains several waste recipes). It is noteworthy that the relatively large differences in EC between percolate and runoff were generally not reflected as strongly in the release of contaminants. This is probably due to pH-dependent (solubility controlled) leaching of these elements, which do not significantly contribute to EC. This would imply that a pH-static leaching test could be used to estimate the release under field conditions, rather than just using the results of a tank-leaching test. In addition, the low volume to area ratio in this scenario and the alternating wet and dry periods led to significant delays in the projected release in comparison with the common assumption of a zero boundary condition for diffusion in a tank leaching test. In general, the concentrations of Mo were high whereas Pb, Cu and Zn were relatively low. This would imply that anions are more relevant for emissions than heavy metals in this disposal scenario. The research will be continued to validate these initial results and to check whether the range in observed concentrations will change.
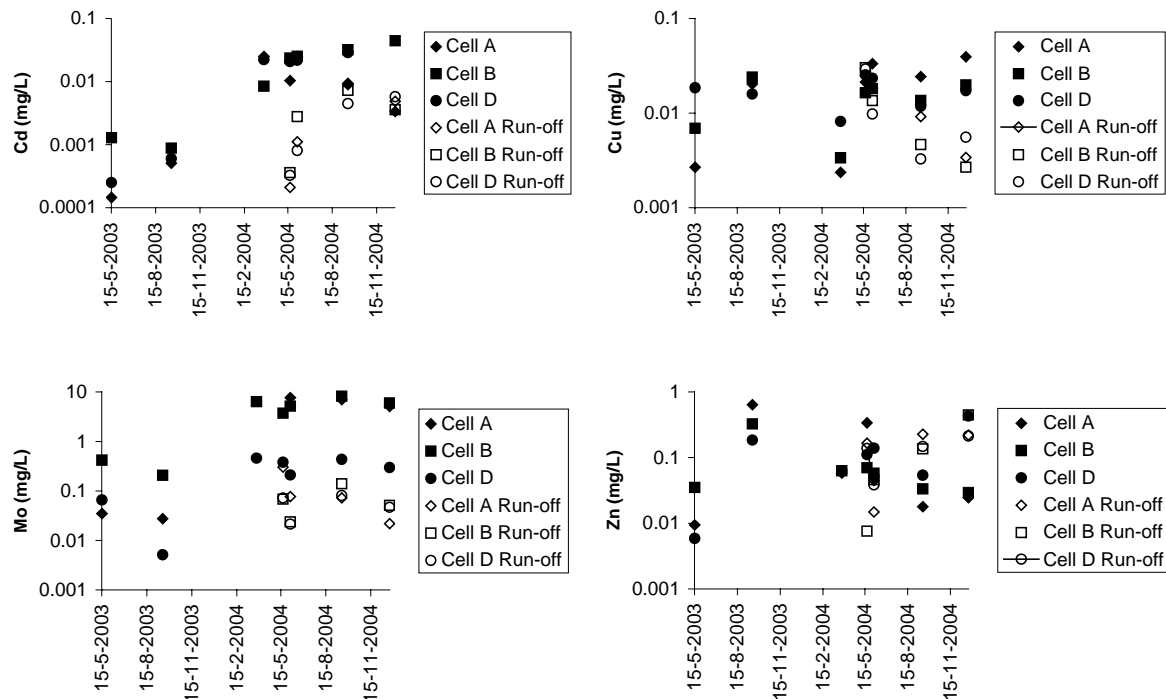
Figure 36   Release of Cd, Cu, Mo and Zn in percolate and runoff from the pilot with a stabilised waste Monolith landfill.

Integration of laboratory, lysimeter and field experiments

The pH-dependent leaching of Cu, Mo, Pb and Zn from stabilised waste in combination with landfill and pilot experiment percolate data, pilot experiment runoff data, landfill core leachate (stabilised waste cores were sampled in the pilot experiment, sliced and leached in the laboratory) and lysimeter data are shown in Figure 37. The solid lines represent the leaching behaviour calculated by ORCHESTRA. The observed leaching behaviour from pH-static experiments can be well described for Cu, Pb and Zn and is adequate for Mo in the high pH range. Mo leaching is not described adequately at neutral to low pH as the model prediction curve deviates substantially from the measured leaching behaviour. In general, the data from landfill core leachate, lysimeter experiments, regular landfill percolate water and the percolate and runoff from the pilot (Figure 37) show a reasonably consistent pH-dependent leaching behaviour when compared with the laboratory data and the model description. Integration of the results from all these different types of testing reveals that the leaching of these contaminants is probably controlled by the same chemical processes, e.g. solubility control by mineral phases, sorption onto HFO and complexation to organic matter. This is also supported by the observation that the relatively large differences in EC between the percolate and runoff were generally not reflected as strongly in the release of contaminants, probably due to the pH-dependent (solubility controlled) leaching of these elements (which do not significantly contribute to EC). This implies that a pH-static leaching test should be used in combination with the results from a tank-leaching test to estimate the release under field conditions. In general, the concentrations of Mo were high whereas Pb, Cu and Zn were relatively low (Figure 37). The monitoring in part of the pilot will be continued to validate these initial results and to check whether the range in observed concentrations will change. Currently, preparations are being made to dismantle the pilot to sample different parts of the exposed stabilised waste and the underlying soil layer. These experiments are needed to verify the

observed results. It is anticipated that the results from this part of the project will be available in the summer of 2006.
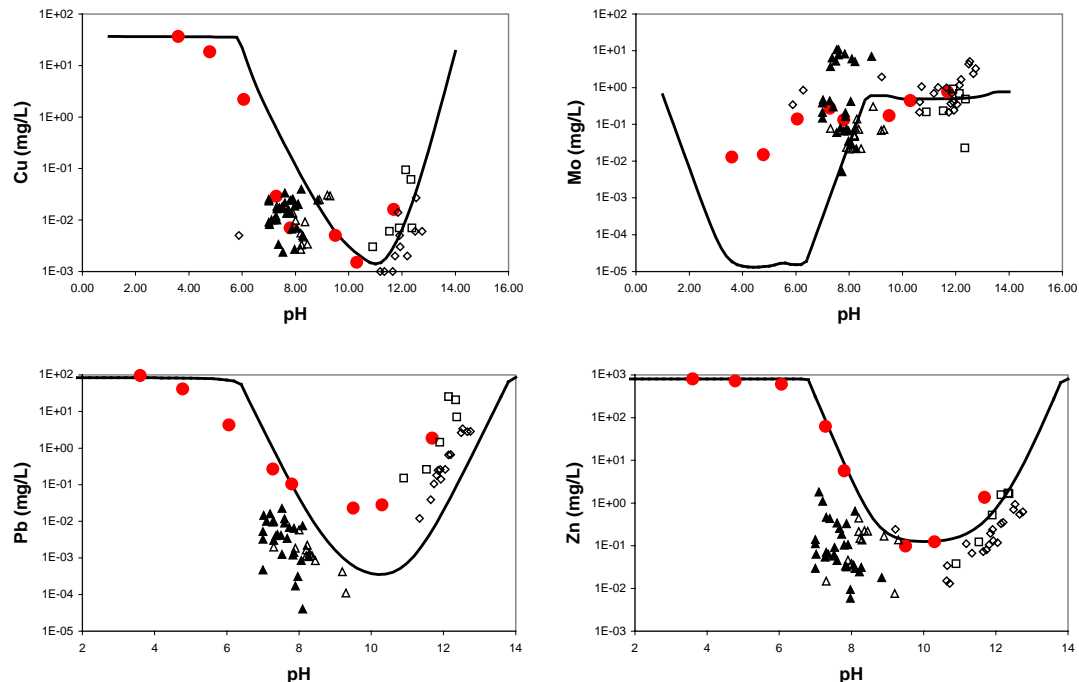
Figure 37   pH-dependent leaching of Cu, Mo, Pb and Zn from stabilised waste (solid circles) in combination with landfill and pilot percolate data (solid triangles), pilot runoff data (open triangles), landfill core leachate (squares) and lysimeter data (diamonds). The solid line represents the predicted leaching behaviour by geochemical modelling (*10*).

Scenario description

During the pilot, the important physical and chemical processes were studied to make a realistic scenario description for this type of landfill. Water balance is important and was quite different from that usually observed for other types of landfills. Calculations on both the whole landfill compartment and the pilot experiment showed that only about 10 to 20% of the total precipitation is measured as percolate or runoff. This result is surprising and suggests that evaporation could be a dominant factor. It must be realised that additional measurements are needed to validate these results before conclusions can be drawn on the percolate water production.

Another process that plays an important role is carbonation. The highly alkaline waste material acts as a CO_2 pump and neutralises the outer layer. Analysis of core samples from 0 to 60 cm have indeed indicated that the outer 2 centimetres of the waste material is carbonated after 4 months (*10*). Fitch and Cheeseman (*6*) have identified $CaCO_3$ minerals in environmentally exposed stabilised waste. Formation of new minerals in the outer layer can seal the pores of the waste material, leading to a reduced diffusion speed and a subsequent lower release of contaminants. It should be noted that the magnitude of this process is not yet known.

The pH in the percolate water is buffered by the presence of the soil layer at the bottom of the waste compartment. pH measurements revealing a neutral pH in the percolate water have

confirmed this mechanism. In an earlier study it was estimated that the soil layer can neutralise the alkaline percolate water for a period of 55 to 115 years; this was based on a constant infiltration rate of 300 mm/yr (*35*). This implies that a soil layer below the stabilised waste can be an important design criterion for this type of landfill.

It is envisaged that the release of soluble salts is predominantly controlled by diffusion. However, the strong pH-dependent leaching of many heavy metals indicates that solubility control by mineral phases might be the dominant release mechanism for these contaminants. These observations imply that it is too simplistic to estimate the long-term release based solely on an extrapolation of the diffusion processes.

Regulatory aspects

The regulatory framework for stabilised waste landfills is still in development. When the criteria for the different landfill classes were set in Annex II of the Landfill Directive (*2*) it proved to be impossible to derive criteria for stabilised monolithic waste due to a lack of information on release and release controlling factors in stabilised waste monofills. Both at the national and the EU level it was decided that additional information is needed to develop meaningful criteria for this type of landfill. Therefore, regulatory controls for this type of landfill are left to the individual Member States until such time as EU-wide criteria are established.

The Landfill Directive was taken as a starting point against which to assess the emissions from the pilot. Limit values were calculated using a so-called point of compliance (POC1). This approach is consistent with the methods used for the development of acceptance criteria for granular waste materials. It is currently impossible to model all chemical and physical processes to obtain a 'source term' for monolithic waste. Therefore, the measured concentrations (expressed as 95 percentile values) were taken as the starting point. It should be noted that this approach only reflects the current emissions. It is unclear whether this approach is representative of the long-term emissions; integral modelling is needed to determine this.

Table 9 shows a comparison of measured concentrations in the percolate water in the pilot with calculated limit concentrations at the point of compliance (POC1). The concentrations of Br^-, Cd, Cl^-, Cr, Mo, Sb, Se and SO_4^{2-} exceed the calculated limit values for inert waste. The concentrations of Br^-, Cl^-, Mo, Se and SO_4^{2-} also exceed the non-hazardous waste limit values. The concentrations of these components also exceed the hazardous waste limits, except for Se.

Table 9   *Comparison of measured concentrations (n = 28) in percolate water in the pilot with calculated limit concentrations at the point of compliance (POC1). Limit values are calculated for landfills that accept hazardous, non-hazardous and inert waste materials (respectively). The ratio of measured concentrations to the inert limit values are calculated. Values in red show that the '95-percentile concentration' exceeds the stated limit value.*

Element	Average	Stand. Deviation	95 percentile	Hazardous	Non-Hazardous	Inert	Ratio
	(mg/l)	(mg/l)	(mg/l)*	(mg/l)	(mg/l)	(mg/l)	95perc/inert
As	0.012	0.01	0.041	2.83	1.00	0.05	0.82
Ba	0.334	0.31	1.022	16.38	11.24	2.00	0.51
Br	446.449	641.53	2,001.100	0.32	0.31	0.31	6,455.16
Cd	0.015	0.02	0.040	0.48	0.19	0.004	10.02
Cl^-	23,058.530	25,693.14	81,455.425	3,150.00	3,139.00	80.00	1,018.19
Cr	0.051	0.08	0.210	27.63	9.39	0.05	4.20
Cu	0.017	0.01	0.031	4.41	1.85	0.20	0.15
F^-	0.405	0.21	0.703	35.10	24.09	1	0.70
Hg	0.00005	0.00004	0.0001	0.55	0.19	0.001	0.09
Mo	2.637	3.63	9.816	4.66	2.11	0.05	196.32
Ni	0.019	0.02	0.039	5.65	2.00	0.04	0.96
Pb	0.004	0.01	0.013	2.83	1.00	0.05	0.27
Sb	0.003	0.02	0.013	0.20	0.11	0.006	2.23
Se	0.104	0.13	0.372	0.40	0.21	0.01	37.20
Sn	0.001	0.005	0.010	2.41	0.95	0.77	0.01
SO_4^{2-}	2,772.092	2,966.58	8,634.675	3,150.00	3,139.00	100.00	86.35
V	0.002	0.03	0.002	3.96	2.13	0.37	0.01
Zn	0.121	0.15	0.398	17.46	6.51	0.40	1.00

*   The '95-percentile concentration' is the concentration for which 95% of the values are lower than this concentration.

More information on the research related to monolithic control is presented in the research report Monolith (*36*), which is also referenced in section 6.3.

5.   Evaluation

5.1   Introduction

The outcome of this work will lead to the formulation of design rules and operational procedures for a landfill in which, within one generation, concentrations of pollutants in its leachate are reduced to a level that an emission of such leachate causes no adverse effects to the soil or groundwater. Atmospheric emissions from such a landfill should also be negligible.

The identified processes that control the release of a specific component are determined by:
1   The composition of the mixed waste input (waste selection).
2   The application of a specific pre-treatment to reduce leaching of components (immobilisation).
3   The occurrence and extent of biodegradation of organic material (biodegradation).
4   The solubility of various mineral and chemical forms in which components can be present (solubility control).
5   The hydrological conditions that the waste is exposed to (flushing).

The typical emissions from a landfill are presented in Table 10, alongside the mechanisms/processes that can be applied to mitigate their influence. A distinction is made between, organic macro components (COD, BOD, N_{kj}), organic micro components (e.g. PAH's, solvents), metals, oxyanions and salts.

Table 10   *Combination of processes and types of emission components*

Process / Component	Waste-Selection	Solidification/ stabilisation	Biodegradation	Solubility Control	Flushing
Org macro components	X	-	X	X	x
Org micro components	X**	X	x	X##	X#
Metals	X	X	X	X	x
Oxyanions	X	X	x	X	X
Salts	X	X	-	-	X

Legend:
**   Evaluation by leaching is still lacking.
#   only for water soluble organic contaminants
relevant for poorly water soluble constituents (DOC reduction will help reduce release)
An 'X' in the table indicates that the component is strongly influenced by the process concerned. As such this means that the process can be applied to minimise or control the release of the component.
An 'x' indicates that the process has some effect on the component, but this may be a side-effect of the main effect that can be achieved for other components;

Table 10 shows that there is no 'one-to-one' relationship between the process occurring and the components released. Components are influenced by a number of processes that cannot be considered individually, as they occur simultaneously. It also shows that for each of the components at least one process can be utilised to reduce the emission potential. These processes were described in detail in Chapter 2.

Based on a general scheme such as that presented in Table 10, a number of design and operational variations can be developed, of which the pilots in the described project are examples.

In the following section a number of subjects will be discussed in more detail. They have been grouped according to an aspect of the project in order to improve the accessibility of this information.

5.2 Regulatory framework

Liquid phase

Criteria for disposal have been establsihed at the European level for the release of a selection of inorganic constituents and a very limited set of organic contaminants. These criteria are presented in Annex II of the Landfill Directive.

Annex II defines the procedures to be applied for the acceptance of waste at the different categories of landfills, namely: landfills for hazardous, non-hazardous and inert waste. A number of waste materials can be accepted without testing at each of these types of landfill. Leaching behaviour is the determining factor for other wastes being assessed against the regulatory criteria. Three types of test are used to set standards for leaching:
- A batch-leaching test at L/S ratio of 2.
- A batch-leaching test at L/S ratio of 10.
- A column leaching (percolation) test.

The limit values are expressed as maximum leachable amounts in mg/kg dry substance for the first two tests. In the column test the value is determined as the concentration in the first eluate produced in the test (at an L/S ratio of 0.1), expressed as mg/l.

The set of criteria for inert waste are of specific interest in the framework of this project. A landfill for inert waste has no requirements for containment and aftercare. This implies that the leachate produced is considered harmless to the environment. The conditions of the inert waste landfill do not necessarily have to be met from the beginning of landfilling (which would be necessary from the current regulatory perspective), but may be defined for this project as a condition to be reached when the active period of leachate treatment has expired. This implies that management in the 'active' period can help to ensure that the desired end condition is reached. Such active management may include a more stringent acceptance policy to exclude waste streams, which possibly meeting current Annex II specifications, but that definitely will affect the leachate quality disproportionally. Allowing infiltration or active leachate recirculation may prove necessary to reach this final storage condition.

Controlling and managing the release behaviour of a landfill or landfill cell to reach inert waste conditions appears to be technically feasible for many, if not all of the constituents specified in Annex II. This, however, does not guarantee that a sustainable landfill concept will be acceptable in the long-term. It is necessary also to consider currently non-regulated substances (e.g NH_4^+) to ensure acceptance. Such a broad evaluation has been made in this work, allowing assessment well beyond the current Annex II criteria. There are no criteria based on release for organic contaminants (except phenol), but rather they are based on composition. Assessment of such contaminants should, as for inorganic elements, be based on leaching, as only a minor fraction of the total content is leachable. The test methods are in place to be able to do this, which implies that during the revision of the Landfill Directive, this

aspect can be addressed more appropriately. Further work is needed to define a protocol to assess whether the condition of maturity, corresponding to inert waste behaviour, has been reached after completion and closure of the landfill.

Gaseous phase

At present, guidelines setting out maximum landfill gas emissions to the atmosphere are already effective in countries such as UK, France and Finland. Other Member States are still developing their own. However, there is no agreement at the European level on both methodology and the maximum acceptable levels of emissions.

It is not made less complicated by the fact that landfill gas emissions have a broad range of possible effects on health, safety, nuisance and the environment (including global warming). Bour *et al.* (*37*) approach this complex subject from the concept of trigger values: emission levels above which action should be taken. The conclusion of Bour *et al.* (*37*) is that the risks to health from landfill gas are very low. Therefore, he proposes trigger values on the basis of the methane production rate, to minimise the impact on global warming, Different options are available for determining a trigger value for methane production, namely:

- Based on natural attenuation. Topsoils of landfills are able to break down the methane at a maximum rate of about 1 dm^3 CH_4/m^2h (e.g. (*26*) and others). The methane flux itself might be a basis for defining the maximum methane flux allowed. Bour *et al.* (*37*) propose a two step approach in which levels above 1 dm^3 CH_4/m^2h require active collection, below 1 l /m^2h a simplified risk assessment is needed and at levels below 0.1 dm^3 CH_4/m^2h natural attenuation is assumed to reduce methane emissions to acceptable levels. Others simply propose the 1 dm^3 CH_4/m^2h as a methane emission limit (*38,39*). Such an emission limit may also be used to derive a minimum degree of biodegradation, assuming a formation model for a standard landfill, e.g. a conventional landfill 10 metres high theoretically reaches a methane formation of 1 dm^3 CH_4/m^2h after about 30 years, when about 95% of the methane potential has vanished (assuming a half-life of 7 years and a methane potential of 130 m^3 STP per m^3 of waste). However, in practice emissions at most conventional landfills are already below 1 dm^3 CH_4/m^2h about 5 years after closure (*40*). It is evident from both theory and practical experiences that landfill bioreactors with effective enhanced biodegradation, will meet the threshold value of 1 dm^3 CH_4/m^2h.
- Based on the feasibility of landfill gas extraction, e.g. UK Environment Agency (*41*). A comparison of the projection for landfill gas formation with a trigger value of 50 to 100 m^3 STP CH_4/h, which is the smallest scale on which extraction and flaring is considered technically feasible.
- Based on the 'shadow' costs of emissions. 'Shadow' costs are based on the maximum costs accepted by society to mitigate similar emissions from other sectors. One tonne of waste produces approximately 50 kg of methane. This has the same impact on climate change as 1 tonne of CO_2 (the global warming potential of CH_4 is about 21 (*42*)). Under the Kyoto protocol, the right to emit CO_2 can be bought and sold and trading prices are currently well below €10 per tonne. A maximum remaining methane potential might be related to the costs that are accepted to mitigate similar amounts of greenhouse gases from other sources, e.g. a residual methane potential of 5% (95% stabilisation) gives a potential emission of 0.05 tonne of CO_2 or shadow costs for emissions of €0.50 per tonne of waste, assuming the upper limit of current CO_2 trading prices of €10 per tonne.

Selection and definition of guidelines for maximum methane emissions from landfills is a task for legislators. The limits suggested above seem to be relatively easily met for the concepts studied within the sustainable landfill programme:

- For the predominantly inorganic waste landfill, the methane release falls well below the criterion of 1 dm^3 CH_4/m^2h, due to the lack of biodegradable components.
- In the organic waste cell the biological degradation was shown to be stimulated in such a way that the remaining gas production was below the criteria mentioned above after a period of four years: emissions were measured to be negligible: less than 0.1 dm^3 CH_4/m^2h (*43*). In other situations the height of the landfill and the actual concentration of organic material will determine when sufficiently low levels can be reached.
- If methane emissions remain unacceptably high in specific cases, fallback options exist to reduce emissions further, e.g. aerating the waste in the build up or final phases of the landfill or the addition of a methane oxidising top cover.

In conclusion the low methane emissions arising when landfilled waste is increasingly biologically-stabilised, the availability of fallback options when stabilisation is not as fast as hoped for, and the relatively low impact of any remaining methane (e.g. when expressed as shadow costs) indicate that methane emissions in practical applications should not be the limiting factor for various concepts of sustainable landfill.

5.3   Viability of the approach

Data

Leachate data have been collated from a wide variety of landfills, providing a point of reference for landfill leachate quality as a starting point for improving the understanding of leaching, and incorporating this knowledge into improved regulations. An important outcome of this project is the recognition that there is a need to analyse the major components, as they control the behaviour of regulated substances (trace elements).

Testing approach

The integration of three different levels of testing (laboratory, lysimeter and pilot scale) for the three different sustainable landfill concepts proved to be invaluable and provides an adequate insight into laboratory-field relationships, given that each level of testing has its own advantages and limitations.

The general consistency of the results of experiments at different scales of testing implies that this approach is useful for identifying the reliable laboratory-field relationships. This forms the basis for a proper estimation of the long-term emissions from landfills, which is particularly relevant when assessing the need for aftercare.

Database

A uniform way representing data was chosen in order that the results from the different concepts, different scales of testing including field leachate could be compared. A database/expert system was developed to facilitate the handling of large amounts of data. The results of the laboratory leaching tests were evaluated by geochemical modelling in order that the geochemical and biochemical processes taking place in a landfill, which lead to the release of contaminants, can be understood.

Chemical processes

An understanding of the chemical processes occurring in landfills opens the possibility of predicting the leaching behaviour of landfills using a limited set of key parameters of the individual waste materials. This also provides the means to identify possible interfering external influences that could lead to changes in the long-term behaviour of the landfill.

Modelling

Predictions of long-term behaviour can be made with sufficient accuracy through modelling and verification with field observations, to enable decisions on the need and extent of aftercare measures. This approach gives additional management options to control release by evaluating waste properties prior to disposal. This provides the opportunity for waste stream modification prior to submission for acceptance at a landfill, treatment prior to landfilling and controlled treatment during the operational phase of landfilling.

5.4   Enhanced biodegradation

The decay of organic material is at the base of both the formation of biogas and the pollution of leachate with BOD, COD and N_{kj}. Mobilisation through adsorption onto dissolved organic carbon is also the most important mechanism for dissolution of heavy metals in the leachate, even at very low concentrations of organic carbon in the waste. Therefore, understanding and controlling the biodegradation of waste is of the utmost importance in the development of process-based landfill bioreactors.

In this project, available literature was integrated into an overall biochemical model and subsequently combined with a triple porosity hydrological model. This integrated model, as described in more detail in section 3.6, enables the prediction of the long-term concentrations of BOD, COD and N_{kj} in the leachate of a flushing bioreactor under different conditions and was used to extrapolate the results of the Landgraaf pilot into the long-term.

An important result of this model is the evidence that BOD and COD can be reduced through accelerated biodegradation followed by flushing out the remaining pollutants. N_{kj} however is less easily reduced, since it is used as a nutrient by the methanogenic biomass and for a large part ends up in the bacteria cell structure. After methanogenesis has been completed, the methanogenic biomass decays only slowly. This results in a long N_{kj} tail in the leachate that is hard to reduce.

The speed and completeness of biodegradation can be increased either through recirculation of the leachate or the injection of oxygen, enabling aerobic conversion, see Table 11. Most experience with leachate recirculation and recent Dutch full scale trials at Wijster (*43*) and Landgraaf (see chapter 4) show that an effective system of leachate recirculation can result in full biological stabilisation within a few years. However, there is still no complete understanding of how leachate recirculation affects biological degradation. Leachate infiltration results in an increased moisture content of the waste and as all reactions take place in the water phase, this might increase the speed of biodegradation. Another potentially more effective mechanism is the mixing effect of leachate infiltration, as a result of which local conditions for biodegradation are improved. Nonetheless the high rate of biogas production and the rapid decline thereof (see chapter 4) suggest that biological processes are enhanced in a much larger part of the waste.

The integrated model also gives an insight into speed-limiting processes. These are summarised in Table 11. A surprising result is that in none of the cases is biodegradation of organic material the rate-limiting step that determines leachate quality in the longer term. The main issue for bioreactors is not how to increase the rate of biodegradation. Instead it is how to flush out the large quantities of pollutants and how to deal with N_{Kj}.

Table 11   *Impact of three flushing regimes on bioconversion*

Flushing	Conclusions
Normal (~300 mm/ yr)	Bioconversion is fast compared to flushing. Concentrations in leachate on longer terms (> 30 yrs) are completely determined by hydrology (amount and homogeneity)
Accelerated (~1,500 mm/ yr)	Both bioconversion and flushing are enhanced. Concentration on longer terms (> 30 yrs) is determined by hydrology. On a term of 5-20 years, N_{Kj} remains elevated due to slow release from decaying bacteriological biomass
Fast (> 3,000 mm/ yr)	N_{Kj} concentrations on a term of 5-20 years are increased due to slow release from decaying bacteriological biomass

A method of reducing nitrogen emissions is the aeration of the waste itself. The stabilisation of organic matter is enhanced in this way and the long-term biogas production reduced. The information available on this method is limited. Nitrification was not studied in this project.

The short-term impact of aeration could be:
- Heat generation: the aerobic process generates heat which could lead to a significant increase in temperature. This aspect requires further attention. Temperature increase can be one of the indicators of the need to adjust the aeration capacity or input of water.
- Leachate quality: shortly after the start of aeration the concentrations of contaminants in the leachate will rise due to the transport of pore water. Then due to flushing and enhanced biodegradation the DOC and N concentrations will drop.
- Biogas quality: the methane content could drop to 1-5%, leaving CO_2, O_2 and N_2.

Uncertainties with regards to enhanced biodegradation

The largest uncertainty in enhancing biodegradation is how to achieve and guarantee a sufficiently homogeneous moisture movement in such a way that biological processes are stimulated throughout the whole landfill. An improved understanding is required of the impact of the initial waste composition, pre-treatment (e.g. shredding) landfill management (e.g. compaction) and biodegradation on hydraulic permeability in time. Opportunities should also be explored to improve permeability by adding specific materials to the waste, e.g. shredded materials that do not decompose and ensure some openness of the structure.

5.5   Inorganic materials

The Cl^- and SO_4^{2-} emissions did not meet the criteria for an inert waste landfill. However, when an L/S ratio of 1.0 is reached after 30 years a major portion of the leachable Cl^- will have been washed out. This implies that SO_4^{2-} may be the only remaining critical component. It is related to gypsum solubility control, which cannot be changed. The gypsum content can be lowered, but it cannot be eliminated. Ultimately only dilution in groundwater remains a control measure for SO_4^{2-}, as the gypsum is still controlling its release at the time of closure. This aspect is common to almost all landfills.

Currently, there are no criteria specified for organic contaminants. Therefore, these cannot be assessed against regulatory requirements. Leaching of organic contaminants can be controlled and assessed in much the same way as for inorganic contaminants. The release of poorly water soluble organic contaminants is largely determined by DOC (possibly sub-fractions of DOC).

Geochemical speciation modelling was used in this study to identify the important chemical processes that determine leaching from mixed waste materials in the short and long-term. pH-static leaching tests in combination with model predictions of the leaching behaviour and the speciation of contaminants, in both the solid phase and in solution, provide detailed knowledge of the chemical processes occurring in these apparently heterogeneous materials. Significant progress in geochemical modelling has been made, and many elements can be adequately predicted simultaneously by model calculations.

An understanding of the physical/chemical processes in a landfill enables the prediction of the long-term emissions. Furthermore, geochemical modelling can reveal the important contaminant sequestration processes. Once these are identified, it is feasible that landfill managers can control the amount of reactive surfaces (iron/aluminium oxides and stabilised solid organic matter) and the potential leachable amount of contaminants in order to ensure good environmental behaviour of the waste body. The anticipated outcome is a disposal practice with new waste acceptance strategies for mutual interaction that will reduce the contaminant emissions to acceptable levels in the long-term. Prediction of the long-term leaching behaviour of a landfill is particularly relevant for assessing the need for the continuation of aftercare.

5.6 Aspects of immobilisation of hazardous waste

The 95-percentile concentrations of Br^-, Cd, Cl^-, Cr, Mo, Sb, Se and SO_4^{2-} exceed the calculated limits for inert waste. Br^-, Cl^-, SO_4^{2-}, Mo and possibly Se are the critical constituents with respect to the non-hazardous/hazardous waste limits. It should be noted that this approach reflects actual emissions. It is not clear whether this finding can be seen as representative of emissions in the long-term.

Future modelling efforts will focus on the description of the time-dependent 'source term' of stabilised waste, including the effect of carbonation, wet/dry cycles, pore sealing and the binding properties of the soil layer. The potential to perform such an assessment has been increased significantly by the work in this study. Although this approach is complicated and will be based on a number of assumptions, it is expected that the assessment of the environmental effects of landfilling stabilised waste can be properly addressed.

The measured Sb (detection limit is 0.01 mg/l) and Se (detection limit is 0.06 mg/l) concentrations were below the detection limits (for ICP-AES) in about half of the measurements in the pilot study. Concentrations measured close to detection limits are generally not reliable and should not be used to draw conclusions. Supplementary measurements with a more sensitive technique (hydride generation with atomic fluorescence spectrometry, HAFS) should be carried out to determine whether Se is critical with respect to the acceptance criteria for inert waste. However, the reported Sb concentration can be seen as the upper limit for Sb emissions. Supplementary measurements of Se with HAFS would be useful to enhance the accuracy of the comparison to the non-hazardous waste limit. However,

it is anticipated that re-analysis with HAFS will not lead to a different conclusion with respect to the assessment against the inert waste criteria.

The outer waste layer was carbonated and gave rise to a runoff with a more or less neutral pH. Carbonation probably also affects the pH of the leachate. The neutral pH is also beneficial for solubility controlled heavy metal leaching, which is generally low at neutral pH. However, the emission of oxyanions like Mo and Se reach critical values at this pH with respect to the criteria set for hazardous waste landfills. In addition, Br^-, Cl^- and SO_4^{2-} emissions exceeded the limits for hazardous waste.

Following the observations from core analyses that pore sealing may be important, field verification measurements are planned for spring 2006. This additional work will focus on the degree of pore sealing due to carbonation (precipitation of $CaCO_3$) that has occurred. Pore sealing can lower emissions of all constituents by reducing the speed. Preliminary model calculations and results from another pilot study show that this process can be of importance in stabilised waste landfills.

The soil layer used for protecting the bottom liner neutralised the alkaline percolate water (together with carbonation effects) and served to bind heavy metals. This finding can be used as a design criterion for stabilised waste landfills. Although the effects of carbonation and implicitly the effect of the buffering by the soil layer can be clearly seen it has to be borne in mind that they were observed through indirect measurements. Future work is planned to measure these effects directly after dismantling the pilot.

In general, the data from the landfill core leachate, lysimeter experiments, landfill percolate water and the percolate and runoff from the pilot show a reasonably consistent pH-dependent leaching behaviour when compared to the laboratory data and the model description. Integration of the results from all these different types of testing reveals that the leaching of these contaminants is controlled by the same chemical processes, e.g. solubility control by mineral phases, sorption onto HFO and complexation to organic matter. This implies that a pH-static leaching test should be used in combination with the results from a tank leaching test in order to estimate release under field conditions.

The long-term physical stability aspects of the Monolith landfill have only been addressed qualitatively in this work under 'worst case' conditions. Therefore, conclusions on the long-term stability of stabilised waste cannot yet be drawn. However, it was found that the upper 5 to 10 cm of the exposed material tends to deteriorate and shows signs of swelling. These effects did not seem to occur in the compartment that was not exposed to the atmosphere.

6.   Conclusions & Recommendations

6.1   Conclusions

General

It is feasible to design and operate a landfill that can comply with the standards set out in the Landfill Directive for a landfill for inert solid waste using careful selection of the solid waste input and suitable control measures (pre-treatment, immobilisation, biodegradation, solution control and flushing). It is questionable whether compliance with the European Groundwater Directive is possible for the parameters Cl^-, SO_4^{2-} and NH_4^+.

The project has shown that for a 'Sustainable Landfill':
- The final emissions are lower than for ordinary landfills.
- Emission release occurs within a shorter period of time.
- Active control and prediction of emissions is possible.

The black box has been opened. The defining processes have been identified. Full control over the processes requires additional knowledge of some of the essential aspects of internal hydrology.

It has been shown that the fundamental approach applied to the project has been successful. Knowledge of the processes responsible for the undesired emissions will lead to technological design measures being developed that will allow control of the main processes that give rise to these emissions. The experiments on different scales combined with the generic framework for data interpretation (based on fundamental biogeochemical principles such as the thermodynamic relations between the different chemical species of contaminants occurring in the landfill), provide many explanations for the measured emission behaviour.

The data interpretation framework shows that the prediction of release gives adequate to good results for the experiments at different scales and the materials used. Therefore, it is feasible that extrapolations can be made to estimate the long-term behaviour of landfills. The outcome of the combination of the results indicate that they can provide information on different ranges of L/S ratios and that L/S can be considered to be a scaling factor for time. However, a difficult and as yet not very well understood phenomenon, is the consequence of preferential flow in landfills. The data indicate that the importance of preferential flow increases as the scale of the experiment is increased. The means to quantify the extent of the influence of preferential flow over the different scales has yet to be developed.

Modelling promises to be a viable approach to understanding preferential flow and devising technological measures to tailor preferential flow to our needs.

This project has clearly shown that the geochemical reactions occurring in the landfill tend to reduce the mobility of many of the inorganic compounds, as most of the concentrations are solubility controlled. The technological measure necessary to control this aspect is based on knowledge of the capacity for immobilisation. A careful selection of waste materials can influence the capacity. Potentially problematic waste can be counteracted with waste that has

a high immobilisation capacity. Conservative elements remain a problem because preferential flow reduces the amount of flushing. Conservative elements are those elements for which flushing is the only means to reduce their leaching if they do not biodegrade or physically or chemically react to bind to another element or molecule or precipitate. This means that removal prior to landfilling has to be considered in order to meet the specified targets. Site selection close to the sea and a site-specific risk assessment could be a solution for specific salts.

Organic control

Dissolved organic carbon increases the dissolved concentrations of many inorganic compounds and hence control of the DOC level is a means of controlling emissions. Furthermore, a quota system could be introduced to control the emissions of conservative elements and DOC. It has also been shown that stimulated biological degradation can be a fast process that removes a significant amount of the labile organic matter in the landfill and is therefore a process for reducing the DOC levels.

Methane emissions should not be a limiting factor for various concepts of sustainable landfilling, given the low level of methane emissions arising when waste is increasingly biologically stabilised and the relatively low impact of the remaining methane.

In landfills containing organic material, biodegradation is the most important process as it produces, removes, mobilises and immobilises key pollutants.

Integration of largely existing knowledge resulted in simple, yet effective models that enhance the understanding and enable the prediction of the leaching of pollutants determined by biodegradation.

Biodegradation leads to reduced emissions and can be enhanced by leachate recirculation. The flushing of contaminants from the landfill appears to be the limiting factor.

Inorganic control

Emissions of contaminants from predominantly inorganic waste materials meet the Landfill Directive's criteria for the acceptance of inert waste materials, except for Cl^- and SO_4^{2-}.

There is generally a good agreement between the results from the different scales of testing. This implies that the laboratory percolation test is adequate for assessment of the long-term behaviour of waste materials. Differences in results from the various scales of testing can often be related to preferential flow (for mobile constituents) or the differences in the redox situation.

Mixing of waste materials enables the creation of beneficial conditions for the leaching behaviour of contaminants

Oxyanions and salts seem to be critical parameters in the assessment of stabilised waste with respect to the limits for hazardous waste. When the 95 percentile concentrations are assessed against the inert criteria, the elements Br^-, Cd, Cl^-, Cr, Mo, Sb, Se and SO_4^{2-} are critical. It should be noted that this approach reflects the actual emissions arising. It is not clear whether this situation can be seen as representative for the long-term.

Monitoring of the stabilised waste pilot showed that carbonation of the waste material leads to a runoff with a neutral pH. Carbonation of the produced runoff itself could also contribute to the observed neutral pH values.

The soil layer used to protect the bottom liner in the stabilised waste pilot played an important role in neutralisation of the percolate and metal binding. Preliminary calculations show that this soil layer can neutralise alkaline percolate water for a long (at least 55 to 110 years) but not indefinite period. More work is needed to study the effect of carbonation and pore sealing as these processes contribute to the estimated neutralisation time.

6.2   Recommendations

The process of defining technological design measures that enable the control of processes that give rise to emissions would be advanced with the gathering of further mechanistic knowledge of the processes resulting in emissions from landfills.

The regulatory framework provided by the Landfill Directive and its Annex II, requires further adaptation to allow for the assessment of mixtures of waste. The requirements defined for landfills for inert waste provide a good basis for a first evaluation of sustainable principles for landfills.

The inert waste criteria do not necessarily have to be met from the start of the landfilling activities, but may be defined as a condition to be reached when the active period of leachate treatment has expired. More research on the influence of operational management would help to ensure the desired end condition is reached.

Additional information is needed both at the national and EU level to develop proper criteria for stabilised monolithic waste.

pH-dependent leaching tests in combination with tank leaching tests may be adequate for estimating release for monolithic materials under field conditions.

The water balance in the Monolith concept should be studied in more detail in order to validate the observed results.

The effect of carbonation and pore sealing need to be studied quantitatively, as these processes contribute to the estimated neutralisation time of the protective buffering soil layer that was used in the Monolith concept.

Research on preferential flow with remote sensing techniques is planned by TNO-MEP.

6.3   Further information

Full coverage of the technical and detailed information generated during this project (Sustainable Landfilling 2000-2005) is presented in four separate reports:
- Bioreactor research ((*29*), supported by Grontmij and TNO-MEP).
- Equifill research ((*20*), supported by ECN).
- Monofill research ((*36*), supported by ECN).
- Database and modelling ((*21*), supported by ECN).

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More information can be obtained from the Dutch Sustainable Landfill Foundation. Please contact the Dutch Sustainable Landfill Foundation tel. + 31 73 6279444, www.sustainablelandfilling.com or www.duurzaamstorten.nl.

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