Reduction of the long-term emission potential of existing landfills

Final Report Phase 2

Dutch Sustainable Landfill Foundation

Colophon

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1. Introduction

Landfilling has shown a significant shift from a strong emphasis on isolation technologies towards obtaining a fundamental understanding of processes occurring in landfill bodies. One of the main reasons for this shift in thinking is the realization that isolation technologies alone cannot guarantee long-term protection of the environment. In addition we more and more realize that in order to have an equally prosperous society in the future it is necessary to find ways of sustainable development. The fundamental idea of sustainability is not to burden future generations with problems created by previous generations. Current regulation of landfills removes environmental risk by requiring a complete sealing of the landfill with impermeable geo-membranes. These membranes prevent water from infiltrating in landfill, and as a result the driving force for leachate emission is removed. A major drawback of this approach is that the entombed landfill continues to have a significant emission potential which will be activated whenever the geotechnical measures to prevent water from infiltration eventually fail. The Dutch ministry of Housing, Spatial Planning and the Environment has recently acknowledged this fact by stating that sustainable landfilling is an approach that should be aimed for (LAP2, National Waste management plan).

Considering the issue of sustainable development, landfills pose a significant challenge. The so-called contaminating life span of a landfill body [54] is defined as the period a landfill body is capable of producing emissions (both gas and liquid) in which substances are present at levels that could have an unacceptable impact on the surrounding environment. Now we realize that the contaminating lifespan of modern landfills may last for centuries [14, 20]. This clearly is not sustainable as future generations are burdened with more or less eternal aftercare of modern landfills. The current approaches in some countries, where funds are provided for this eternal after-care, assume that our current day financial and regulatory institutions will survive for centuries. One can have doubts about this assumption as very few institutions have been able to survive for such a period of time, some examples of exceptions are the Bank of England [20] and the Dutch Water Authorities which have survived for more than 300 years.

The current approach of releasing a landfill from after-care based on monitoring data over a certain period after closure is not a sound way of assessing the risk as time alone is not the relevant parameter. Instead, knowledge of the rate of the processes responsible for stabilization of the landfill body is essential in order to determine if risks of unwanted emission are low enough to consider release from (extensive) after-care.

1.1 Dutch Foundation for Sustainable Landfilling (DFSL)

The DFSL was founded in order to stimulate the development of alternative landfilling concepts based on sustainability principles. Background information on the goals and projects carried out within the framework of the foundation can be found on the internet (http://www.duurzaamstorten.nl/wawcs0122289/ln-home-en.html). The definition of sustainability used here is that the consequences of an action do not last longer than a single generation. A generation is taken to be 30 years.

The main concept behind the initiatives of the DFSL is to work with natural processes instead of against them. Natural processes occur within the landfill body and our approach is to let nature do part of the work for us. For reducing the after-care burden of existing landfills this means that various natural processes, such as leaching and the natural decomposition of organic matter, need to be stimulated so that the conditions within the landfill will be more in equilibrium with the surrounding environment.

Different technological options are available to the landfill operator for controlling the long term emission potential. In the previous phase on the DFSL the following options were investigated in order to create new more sustainable landfills:

- Pre-treatment
- Biodegradation
- Immobilization
- Solubility control
- Flushing

In this project additional technological options are added to this list in order to reduce the long-term emission potential of existing landfills.

1.2 Goal and ambition of report

This report gives a short summary of the current state of knowledge, the results available from the different projects carried out within DFSL and important findings from the international literature. The goal of this report is to present an integrated overview so that it becomes clear how future projects and research can be optimally focused in order to achieve the goals of the DFSL as fast as possible.

This report is the final report of the project Core Team, Sustainable Landfilling Phase II. The main products of this phase within the DFSL were the feasibility studies for the two demonstration projects at the Kragge and Wieringermeer landfills [78-82].

The remainder of this chapter will give a short introduction where we try to link the concept of reducing the long-term emission potential to risks associated with landfills, the processes occurring within the landfill and the technology available for manipulating these processes.

Chapter 2 introduces the recently developed concept of Sustainability Potential Analysis (SPA) as a tool to including landfilling in to the wider context of sustainable development. Principles of SPA provide a good foundation for quantitative evaluation of risk associated with landfills. Chapter 3 gives an overview of the current state of mechanistic knowledge on processes occurring in the waste body and how these processes are related to the long-term emission potential. How to manipulate and control these processes using landfill technology is described in chapter 0. Chapter 5 gives short summaries of the progress in the different projects which are running within the framework of the DFSL. Finally conclusions and recommendations are given in chapter 6.

1.3 Sustainable landfilling and emission potential

The main aim of the DFSL is to develop and implement concepts which reduce the long term threat of landfills. Therefore we aim to reduce the contaminating life span of a landfill body [54]. In order for this approach to be successful it is necessary that the remaining risk

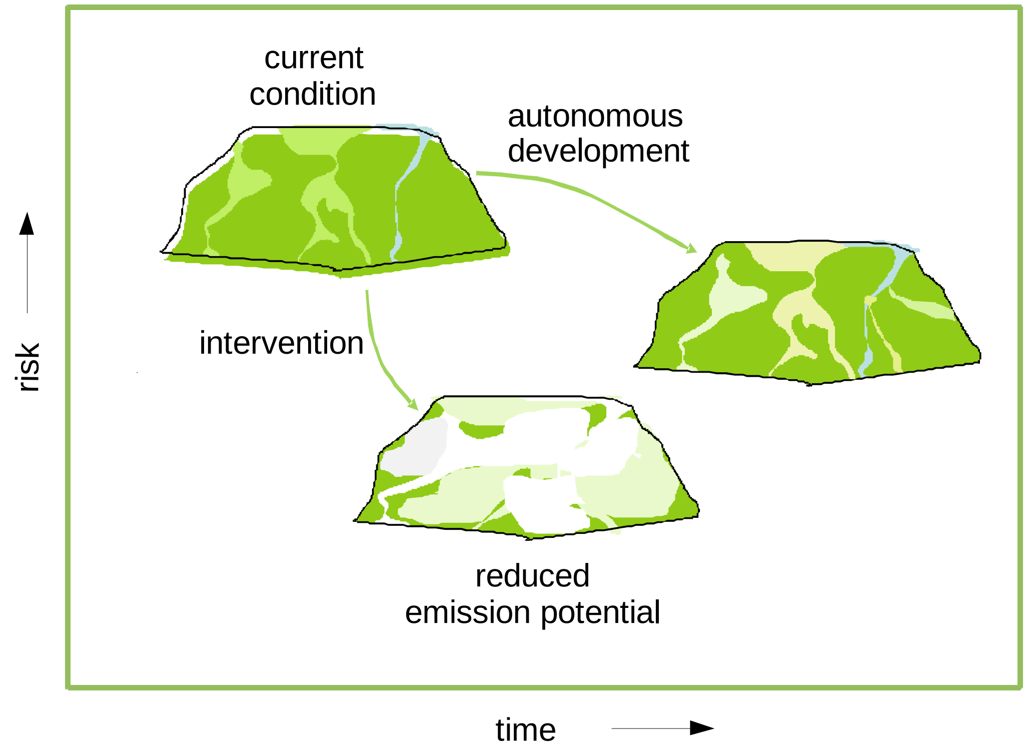

Figure 1.1 Conceptual model for reducing emission potential of a landfill

associated with the presence of the landfill is quantified. An important factor in this respect is the long-term emission potential of a landfill.

Within the research group, a conceptual model is developed, depicted in figure 1.1. The assumption is that stimulation of the in-situ bio-degradation of organic material in the landfill body will result in geo-chemical and physical stabilization of the waste and therefore, in a significant reduction of the long-term emission potential.

Currently, more and more evidence is being gathered showing that the geochemical stability of waste is dominated by the speciation of organic matter in the landfill body and that enhanced biodegradation of the organic fraction in the waste leads to reduced emission levels. Most data for this assumption has been obtained from laboratory and intermediate scale experiments. Data obtained from full-scale experiments is much less conclusive. Benson et al. [6] reviewed data from five landfills in the USA which were operated as a bioreactor landfill and compared the data with conventional non-bioreactor landfills. The authors were not able to draw a definitive conclusion that leachate recirculation leads to accelerated waste degradation. Results indicated that the amount of leachate recirculated was perhaps too little. The conclusion was that more detailed monitoring is required at full-scale bioreactor landfills.

1.4 Risk, processes & technology

1.4.1 Risk

Initially the concept adopted in this report is based on the idea that risk is defined by the probability of an event occurring multiplied by the impact of this event occurring.

Regulation for landfills is currently focused on managing the long-term risk associated with the presence of the landfill for humanity, the surrounding eco-system and groundwater. Understanding risks associated with landfills is an extremely complex field. Current regulation is based on the precautionary principle paraphrased as "better safe than sorry". This has resulted in the eternal entombment of landfills in water tight geo-membranes.

Long-term risks associated with a landfill are, however, primarily related to emissions of contaminants to the soil and groundwater after protective measures have failed. This risk of groundwater pollution is probably the most severe environmental impact from landfills because of the longevity of the emission potential within landfills. Assuming that geotechnical systems will always eventually fail, the probability of emissions occurring from the landfill is certain. Adding to this the fact that the emission potential of entombed landfills remains high during entombment the impact on the groundwater will be high. To make things even worse, failure occurs in the future so the burden of our waste is shifted to next generations. However, we also must realize that failure of the lining will occur gradually, so the emission potential will be released slowly. Perhaps even so slow, that no risk will occur.

Adopting an approach where reduction of the emission potential leads to a significant decrease in impact is the best approach to reduce the long-term risk. Methods for quantifying the risks under different scenarios still have to be developed. Key elements of such risk assessment methods are:

- landfill processes (related to management and quantification of emission potential);
- landfill technology (related to controlling emission potential);
- emission as a function of time;
  - role of the stagnant zone in release at long term of mobile constituents (salts, NH_4, DOC bound species);
  - change in behavior of released compounds due to changes in geochemical environment as leachate moves out of the landfill.

However, a sustainability assessment should indicate more aspects than environmental risk alone. In chapter 2 we present the concept of Sustainability Potential Analysis of landfills [32], which is an approach that broadens the concept of risk analysis with other aspects related to the capacity of a landfill to hinder or even support sustainable development.

1.4.2 Risk according to the IWWG-SLTG

The Sustainable Landfilling Task Group of the International Waste Working Group (IWWG-SLTG workshop report 07-10-09) held a workshop at the Sardinia 2009 Symposium. More than 70 participants engaged In a discussion trying to define sustainable landfill.

Most participants present agreed to adopt a pragmatic approach in which it is acknowledged that landfills are a reality, will be among us for a long time and that we have to do the best we can to reduce their emissions. It was concluded that we should emphasize striving for acceptable risk rather than achieving sustainability. Sustainability is not a clear concept and in relation to landfill it causes opposition among regulators and the general public. Moreover once it is sustainable, can it still be called a landfill? Or should it be considered a harmless part of the earth? It was suggested that we are trying to define a desired ‘end-point’.

It was concluded that the landfill industry and the regulators essentially need a definition in order to agree on certificates of completion. Completion is the moment at which the responsibility for remaining risk is transferred from the operator to society. For operators, regulators and society this is a very important moment. The key issue is more about risk and risk assessment than about sustainability. It was agreed this should be considered in the definition.

An issue of some importance is the concept of ‘undisturbed contents of the landfill’. Nature could disturb the contents. If on the other hand all potential disturbances have to be considered, a target may become impossible to meet. For instance under specific pH and redox conditions various heavy metals are not mobilized. In case mobilization due to changing redox conditions has to be accounted for, more effort will have to be made to remove the heavy metals from the waste body. It was agreed to remove ‘undisturbed contents’ from the definition. Therefore, disturbance will have to be included in the risk assessment.

Risk can only be assessed for a specified use and not for all likely after-use purposes. Consequently the after-use has to be decided before the risk assessment can be carried out. A definition of acceptable risk is also needed to provide guidance for after-use and the remaining minimal (custodial) care during after-use. In Japan functional stability includes the ability to use the landfill for other purposes. It was agreed that functional stability cannot be separated from the surrounding environment and from the proposed after-use. Therefore in the risk assessment they have to be considered together.

Considering these issues the workshop concluded on the following definition (or framework) of acceptable risk for landfills in the context of aftercare completion:
- The landfill reaches functional stability (based on site-specific physical, chemical, and biological characteristics of the waste mass and its location) such that the landfill, taking into account its proposed after-use, is unlikely to pose an unacceptable risk to human health or the environment;
- During the process towards stability no unacceptable risk should occur;
- This situation should be reached as quickly as possible and within the financial provision time;
- The funding for completion of aftercare has been secured and allows for appropriate after-use of the site with minimal (custodial) care.

1.4.3 Landfill processes (a conceptual model)

In order to describe what we know of the different processes occurring in a landfill body we use a conceptual model of the physical, chemical and biological processes occurring in the landfill. Our concept is based on the fact that the landfill body consists of three phases:
- a solid phase;
- a liquid phase;
- a gas phase.

These three phases are heterogeneously distributed throughout the landfill body and a wide range of complex reactions and interactions occur in and between these different phases. In order to describe the reactions in the heterogeneous landfill body, it is essential to realize that all these processes occur at all scales between the pore-scale and the landfill-scale. The pore-scale processes occur in units, considered to be homogeneous, the so-called representative elementary volume (REV). The landfill-scale processes occur between the different REVs [27]. Processes discussed in report are:

- In-situ biological degradation of organic matter resulting in production of gas and heat and increased concentrations of dissolved organic carbon in the leachate;
- In-situ (bio)geochemical interactions between the different substances in which, depending on pH and redox status, substances will dissolve or form precipitates, form complexes with mineral surfaces and solid organic matter or form soluble ligands with dissolved organic matter;
- Precipitation and irrigation will cause water (with dissolved substances) to flow through the landfill body, eventually producing leachate. The flow results in transport of heat, dissolved gases and substances;
- Due to pressure differences and active suction, landfill gas (with CO_2, CH_4 and in aerated landfills even N_2 and O_2) will flow through the landfill. Gas flow takes place through the gas filled pore-space.

Our conceptual model of a landfill is based on the following ideas and assumptions. The landfill body is a heterogeneous structure which consists of horizontally compacted waste (due to the way a landfill is filled). This structure leads to a system of preferential flow paths, present in 10 to 30 % of the pore space of the landfill body. Water and gas flow through these preferential flow paths cause gradients to develop in the landfill body which drive a range of biogeochemical processes in the landfill body. Controlling water and gas flow in the landfill body provides us with technical means to manipulate the processes occurring within the landfill body, thus giving us the opportunity to drive the landfill to a condition of minimum emission potential.

1.4.4 Technology for management of long-term emission potential

In order to reduce the emission potential of a landfill, technological measures need to be applied in order to enhance and control the naturally occurring processes. In previous research of the Dutch sustainable Landfill Foundation, options for process control are identified and summarized [37]. These options are related to:
- Controlling the composition of the mixed waste input (waste selection);
- Application of specific pre-treatment to reduce leaching of components (heap leaching, immobilization);
- The occurrence and extent of biodegradation of organic material (biodegradation);
- The solubility of various mineral and chemical forms in which components can be present (solubility control);
- The hydrological conditions that the waste is exposed to (flushing);
- The composition of the gas phase in the landfill body (aeration).

2. Sustainable management of landfills

2.1 Sustainable management of landfills and Risk

Waste management in The Netherlands is based on the so-called “waste-management” hierarchy, which prioritizes (1) waste reduction over (2) reuse and recycling over the last two steps (3) incineration and (4) safe disposal in landfills of waste and waste residues. This hierarchy also underlies the EU landfill directive which demands that EU member states reduce landfill of biodegradable waste to 35 % of the 1995 level within the next decade. However, despite this directive and further technological and organizational improvements in waste-management, considerable fractions of certain types of waste and residuals of other types of waste treatment will have to be landfilled. This is also the fact in The Netherlands where a total ban on landfill of combustible waste has almost completely been implemented and where the market has invested so much that currently the country has a surplus in waste incinerator capacity. Table 2.1 gives an overview of the development of the amount of landfilled waste in the Netherlands and it is clear landfilling remains a crucial end-point of waste management.

Table 2.1 *Development of landfilled waste in The Netherlands form 1991 to 2007*

	Nr of landfills in The Netherlands *absolute*	Landfilled waste Mega tonnes	Remaining capacity *million m^3*	*Capacity in preparation million m^3*
1992	72	13,3	65,7	*78,0*
1993	69	13,0	82,3	*46,4*
1994	56	12,2	83,9	*41,8*
1995	46	9,8	80,0	*28,1*
1996	47	8,5	76,0	*17,1*
1997	44	7,4	73,9	*14,2*
1998	41	7,1	69,4	*6,7*
1999	38	7,6	63,9	*6,7*
2000	36	6,5	58,4	*21,9*
2001	33	6,5	56,5	*16,2*
2002	31	5,2	54,2	*16,9*
2003*	31	3,4	51,0	*17,2*
2004*	29	2,6	52,2	*12,9*
2005*	27	3,2	50,2	*12,7*
2006*	23	3,6	48,3	*9,5*
*2007**	*24*	*3,7*	*55,4*	*5,1*

Source: http://www.milieuennatuurcompendium.nl/indicatoren/nl0393-Stortplaatsen.html?i=1-3 and http://www.verenigingafvalbedrijven.nl/downloads/vereniging_afvalbedrijven_jaarbericht2008.pdf

* landfill data for 2003 to 2007 is net waste landfilled, excluding the waste that has been re-used as so-called secondary construction materials.

The ambition of the Dutch government, however, is to include both current as well as future landfills in the total sustainable development framework of society. This ambition requires a careful assessment of the environmental risk, the social cost, etc. Application of concepts from sustainability is essential because landfills as the end-point of waste management shift the burden to specific areas where the impact of the landfill is felt by a limited number of people, thus contradicting the intra-generation equity (but this also holds for the location of airports, heavy industry, highways etc.). More important, however, is the issue where today's waste problems could be transferred to future generations due to the potential long-term reactivity of the waste body, thus contradicting the inter-generation equity.

The assessment should not only focus on the environmental risk of a landfill, but also include a range of criteria related to sustainable development (SD). Therefore, the assessment should aim to answer the following questions:

- What is to be sustained, at what scale and in what form?
- Over what time period and with what level of certainty?
- Through what social processes and with what tradeoffs against goals?
- How can the landfills potential contribution to sustainable development be quantitatively measured?

Answering the first three questions requires a combination of value judgments, world views and consensual knowledge, whereas the fourth depends on general systemic characteristics that determine the developmental potential of the landfill. Within the framework of the DFSL we have mainly been focussing on trying to answer the fourth question, mainly addressing mitigating environmental effects of landfills, thus creating conditions which allow high-value reuse of space. The first three questions will have to be answered in the coming years in order to integrate landfilling in the SD framework of our society. This chapter gives a summary of the so-called Sustainability Potential Analysis (SPA) which is an example of a framework in which such an assessment can be carried out. This framework is also an excellent means for integrating the results obtained within the DFSL and for identifying important knowledge gaps which have to be addressed in the near future. In addition it will allow the coupling of sustainable landfill technology to so-called Life Cycle Analysis (LCA) concepts

2.2 Sustainability Potential Analysis (SPA)

SPA of a landfill is a comprehensive assessment approach that considers three important dimensions in a systemic perspective [31-32]:

- **Function:** addresses the goals & demands imposed on the landfill system by society.
- **Structure:** focuses on the physical and organizational structure of the landfill system within clearly defined boundaries. Spatial and temporal relationships are especially important.
- **Context:** brings entities outside the landfill system boundary in to the assessment. Context includes all relevant external impact factors on the landfill system including the geophysical and ecological and anthropogenic environmental factors such as regional climate, geomorphology, hydrology, infrastructure, regulations, etc.

The challenge for SPA is to address these dimensions but also to identify and quantify the "processes", being the interactions and interdependencies between these three dimensions. The aim of a sustainable landfill is to obtain a balance with respect to the traditional *economic, ecological* and *social aspects* of the system. Three hierarchical levels can be identified for carrying out the SPA assessment,

- Pragmatic legal assessment (based on legal regulations often implementing threshold values);
- Benchmarking (an assessment based on the comparison with best performances of other systems);
- Absolute assessment (quantification of the “objective” potential to hinder or support SD).

Within the framework of the DFSL, the assessment will probably be based on the third type because the legal framework needs to be adjusted so that it enables sustainable development of landfills. Therefore the main challenge at the moment is to identify “objective” criteria with which we can quantify the impact of a landfill on the sustainable development of society as a whole. It should be noted that a LCA should be part of the complete SPA.

2.3 Steps for implementation of SPA of landfills in the Netherlands

The goal of the assessment should be the quantification of the potential that a landfill has to hinder or support sustainable development. The goal should include aspects of environmental risk, energy- and material consumption and financial consequences. In order to achieve the identification of these goals several currently used approaches to assess landfills can be combined. The steps should at least include:

- The definition of a conceptual model of the system under investigation;
- The definition of measurable criteria and algorithms to calculate / interpret these criteria;
- The definition of algorithms to aggregate the criteria in to a final result.

The conceptual model underlying the landfill assessment should integrate: (1) energy/material- and financial flows; (2) agent-networks and driving forces; and (3) systemic interdependencies that enables the selection of the assessment criteria and allows for a weighting of the criteria using quantitative approaches such as MAUT (Multi-attribute Utility Theory). MAUT is a mathematical approach which allows a quantification of the desirability of different approaches and scenario’s and which is based on predefined “objective” criteria and weights. Using this approach it should be possible to evaluate scenarios, amongst which we should include scenarios of different contextual developments.

The measurable criteria should be chosen in such a manner that together with the algorithms used for interpretation, so-called *utility* or *hazard* scores can be quantified which indicate the potential of the landfill system to support or hinder SD.

Finally quantified criteria should be aggregated in order to compare different options with each other. Depending on assessment goals, these interpretations will lead to different types of action.

2.3.1 Six generic criteria of SPA

Lang et al. [32] identify six generic criteria of SPA which at least should be included in a balanced assessment:

- Performance and efficiency of the landfill system with respect to its goals. This includes the function of the landfill such as the capacity to store waste, the capacity to protect the environment for an indefinite period of time, but also goals related to economical issues, etc.;

- Well-structuredness of the landfill system indicating the quality of the system and, if required, its ability to meet given functions and to satisfactorily adjust to changes in function;
- Interdependencies with other systems indicating how the landfill system influences, or is influenced by, other systems;
- Buffer capacity and resilience. Landfill systems are subject to many external and internal impacts that can unsettle the landfill system. How does the landfill system assimilate the effects and return to a steady state while still being able to fulfill its function?
- Ability to accommodate. If the assimilative capacity is exceeded, what type of new steady state will be reached? What are the consequences for the capacity to fulfill the required function and other goals?
- Inter- and Intra generation equity which is one of the essential keys for sustainability, are the costs and the benefits fairly allocated within the present generation and between the present and future generations. It should be stressed that this is an essential factor for ensuring long-term stability and viability of the landfill system.

Each of these criteria is specified according to the characteristics of the landfill system under investigation using so-called Functional Key Variables (FKV). Examples of FKVs used by [31] are the control of pollutant release, economic adequacy, technical and constructional structures, organizational structures, informational structures, embedding in the natural context, embedding in the societal context, resilience to changes of the natural context, repair ability of the constructional structures, exposure of future generations, financing of the after measures, etc. Quantification of these FKVs is clearly a major challenge which requires much consensus amongst at least scientists, regulators and landfill operators.

2.4 SPA and the projects within the Dutch Sustainable Landfill Foundation

Within the DFSL the main focus has been on the different pilots that have been carried out. The consequence has been that our focus has been limited to the technical aspects of the landfill body itself. The context of the landfill has not been a specific subject of research until now, although some effort has been spent on the surroundings in the Vlagheide project and in the NAVOS project. In the first phase of the sustainable landfilling project, some attempts have been spent on the financial benefits of the sustainable landfilling approaches, in this phase we have not made any progress in this respect. Adopting the SPA approach will provide an integrated framework for including this financial analysis and it will also allow for developing insights how the burden of different options is shared between the different parties involved. The consequence of the focus chosen in the DFSL is that for the remainder of this report we focus on the knowledge available on processes occurring within the landfill body.

It is clear that assessing a landfill within the SPA requires an analysis at the scale of the whole landfill system. Therefore goals will be identified for the scale of the complete landfill as well. Much scientific knowledge is available on the physical, chemical and biological processes occurring in landfills but much of this information was obtained on a much smaller scale, e.g. the scale of waste samples in the laboratory. Adopting the goals of SPA will force us to find ways of translating the available (quantitative) knowledge to the scale of the complete landfill in its context so that we can quantitatively perform the SPA assessment.

It is clear that SPA is a tool that is highly suitable for developing new landfill technology and approaches, but it can also be extremely valuable in defining more sustainable means of after-care of the current Dutch landfills which are a mixture of the old conditions and the new.

3. Landfill processes

Chapter 6 of the generic part of the feasibility study for two new landfill pilots within the framework of the DFSL [79], gives a summary of relevant processes in the landfill body with respect to emission reduction technology. In this report we will focus on the knowledge base for predicting and controlling the long-term leachate quality in landfills.

Kjeldsen et al. [28] give a review of the present knowledge on the expected long term behavior of landfill leachate in general. Leachate may be characterized as a water-based solution of dissolved organic matter, inorganic macro components, heavy metals and xenobiotic organic compounds. Decomposition of organic parts of the waste proceeds through a complex of microbiological reactions. Solid components as hemicelluloses, cellulose or cell-walls made out of lipids are hydrolyzed and hydrolysis products are converted to smaller soluble molecules and ultimately converted to biogas. Part of the organic waste is very resistant to microbial conversion and stays in the waste, undergoing slow transformations to even more stable material. This remaining organic fraction has a large storage capacity for e.g. metal ions, thus sequestrating part of the pollution potential of the initial waste.

There is a lot of evidence that water flow through landfills occurs through preferential flow paths [17, 44, 51-53]. However, it is only in the last couple of years that we are beginning to understand the consequences of preferential flow for biodegradation of organic waste and the overall stabilization of landfills.

Micro-organisms live in the water phase as single organisms but more generally as consortia in biofilms. Water is essential for the survival of these organisms as it is the means of transport of nutrients to the micro-organisms and waste products from these consortia of micro-organisms. Experiments showing an increase in biological degradation after addition of water have led to the assumption that traditional landfills are too dry and additional water is required to increase biological degradation which has led to the development of irrigation and leachate recirculation landfill concepts also called bio-reactor landfills. But moisture movement also plays an important role. Completely saturated zones with stagnant water have shown very poor degradation (Wens, Hooge Made, Antwerpen). Landfill bodies are extremely heterogeneous on the scale of cubic meters and as a result zones with high amounts of degraded organic matter will lie next to zones in which organic matter has undergone no degradation at all.

Work carried out in the last decade on understanding the leaching characteristics of a large number of materials including landfills [61-62, 86] shows that leaching is a process that can be understood with multi-component geochemical modeling. In addition it has become clear that the concentration levels in leachate of a large amount of inorganic substances are highly correlated with the concentration of dissolved organic carbon present. Later on in chapter 3.3 which we show that the chemical speciation in the leachate can now be described with geochemical modeling and that modeling and measurement results can be compared with data of a large number of different landfills of different age and composition. This approach provides us with an opportunity to assess the future emission behavior of the landfill [91]. Using the results from laboratory tests on fresh MSW as a starting condition, modeling has been carried out to predict behavior at conditions well beyond those of the lab experiments. An

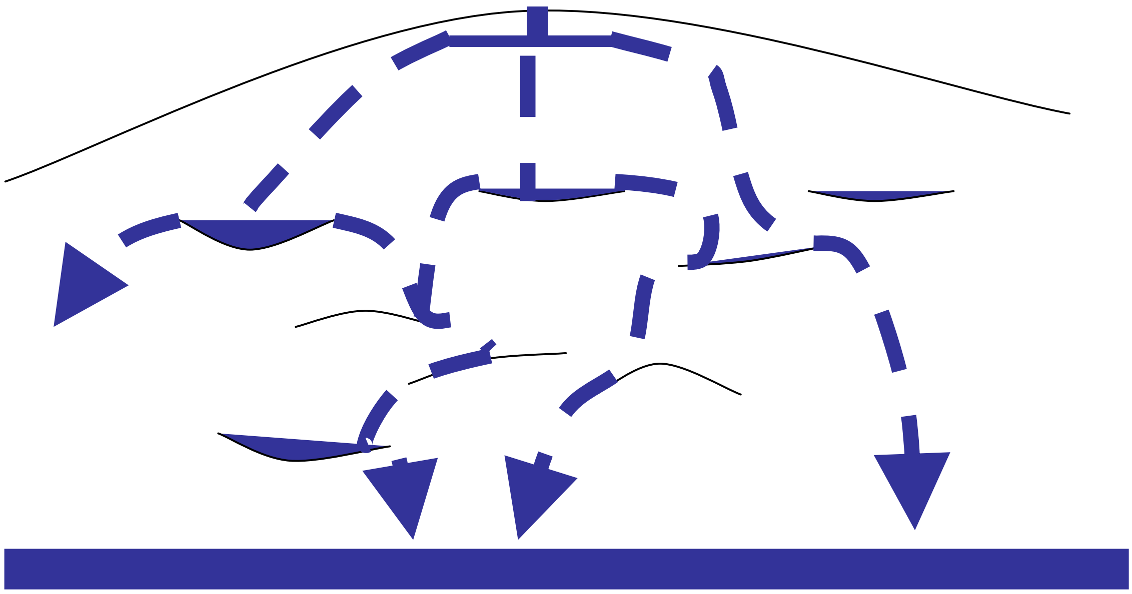

Figure 3.1 Schematic overview of conceptual model of landfill hydrology. The landfill is a heterogeneous structure containing sections which are impermeable for water. Water flow will be diverted and as such a complex flow pattern will develop. Water will flow along these preferential flow paths and only very slowly enter the surrounding waste.

example is the modeling of the effect of biodegradation on the emission by assuming lower redox values in the material (all other parameters remaining the same), different concentration of dissolved organic carbon (DOC) and different solid organic matter content of the waste. These model results have been compared with field leachate data and clearly show that the concentration limits of many inorganic compounds is determined by solubility control [73].

3.1 Flow of water and solutes

Figure 3.1 illustrates the conceptual idea for the flow of water and solutes through the landfill. The results from the different pilots carried out within the framework of the DFSL and abroad indicate that water flow through landfill bodies is highly complex and that the volume of the landfill through which water actively flows can be very small. This preferential flow has major impact on the transport of solutes through the landfill body and on the microbial degradation of organic matter in the landfill.

Control of the flow of water through a landfill body is considered to be the most important factor for stimulating biodegradation and achieving landfill stabilization. However, until now no detailed long term monitoring aimed to unravel the preferential flow of water through the landfills was carried out within the DFSL projects. A number of papers can be found in the literature focused on quantification of preferential flow in landfills and the impact of the flow on solute transport. Little direct information can be found on the impact of preferential flow on microbial degradation.

The rates at which the bio-geochemical reactions occur within a landfill depend on the local conditions [27-28]. Optimal degradation of the organic matter requires (a) presence of sufficient water for the initial hydrolysis of the organic waste into small dissolved organic molecules, (b) neutral pH-conditions, (c) not too high salinity, (d) sufficient presence of nutrients for micro-organisms to grow, and (e) presence of electron-acceptors. These conditions are highly variable throughout the landfill body due to landfilling history, differences in waste type, differences in local landfill topography, different waste ages etc. As a result the bio-geochemical conditions and amount of degradation are heterogeneous throughout the landfill: organic matter can be fully degraded in some parts of the landfill while in other parts of the landfill no degradation has taken place at all (fully dry pockets without any hydrolysis).

Hypothesis for conceptual model Sustainable Landfilling

The main hypothesis underlying our conceptual model is that heterogeneity in degradation in landfill bodies is due to a lack of moving water in the non-degraded zones. When it is too dry, hydrolysis, being the first (and often rate limiting) step for bio-degradation, will not occur. On the other hand, in fully saturated conditions with stagnant leachate, hydrolysis can easily occur but in the first steps of bio-degradation organic acids are produced which reduce the pH. Sometimes accumulation of these acids leads to toxic pH levels for the micro-organisms limiting further bio-degradation. Mobile water in both these cases would reduce the negative effects due to increasing moisture contents, flushing and dilution of the organic acids and the transport of additional nutrients towards the active zones. Therefore, optimal conditions for bio-degradation require both sufficient and moving water throughout the waste body.

Full-scale application of landfill stabilization technology is limited by the fact that it is very difficult to know where and when to apply the right amount of water. Quantification of heterogeneity of the water distribution enables the speed up of landfill stabilization by focusing efforts to zones where degradation is slow. In addition, this knowledge will allow us to make an assessment how far a landfill is stabilized and what potential emissions are to be expected in the future.

Experimental evidence for preferential flow

Johnson et al. [25] present data on a long-term monitoring project in order to unravel the hydrology of the Lostorf MSWI landfill in Switzerland. Different, complementary methods were applied to investigate the flow of water through the landfill: Precipitation and discharge from the landfill was monitored in time, leachate was sampled, electrical conductivity of leachate was measured at high frequency, regular measurements of $^{18}O/^{16}O$ isotope ratios were performed and a number of qualitative tracer studies with fluorescein, pyranine and iodide were carried out during the monitoring period.

The experiments and the results obtained from the measurement campaigns clearly showed evidence for preferential flow which varied throughout the year. Over the complete monitoring period 50 % of the incident rainfall was measured in the drainage, analysis of single rainfall events showed that 90-100 % of the rainfall was expressed in the landfill leachate whereas in summer months this value was 9 - 40 % depending on the rainfall intensity. The response to rainfall was rapid, within 30 to 100 hours 50 % of the water discharged in response to a rainfall event had already left the landfill. The tracer experiments, isotope measurements and the electrical conductivity data showed that 20 - 80 % of the incident rainfall passed directly through the landfill in summer months and around 10 % in the winter months. Clearly, in MSWI ash landfills water flow is highly dependent on the water content. Preferential flow is large under dry conditions, smaller under wet conditions. Care however must be taken when extrapolating these results to other types of landfills which will have very different heterogeneity and structure.

Rosqvist and co-workers [5, 17, 51-53] present results from tracer experiments carried out on waste and landfills at different scales. In the paper by Rosquist and Destouni [52], 55 to 70 % of the water flowed through only 5 to 16 % of an undisturbed waste sample, in the landfill 90 %

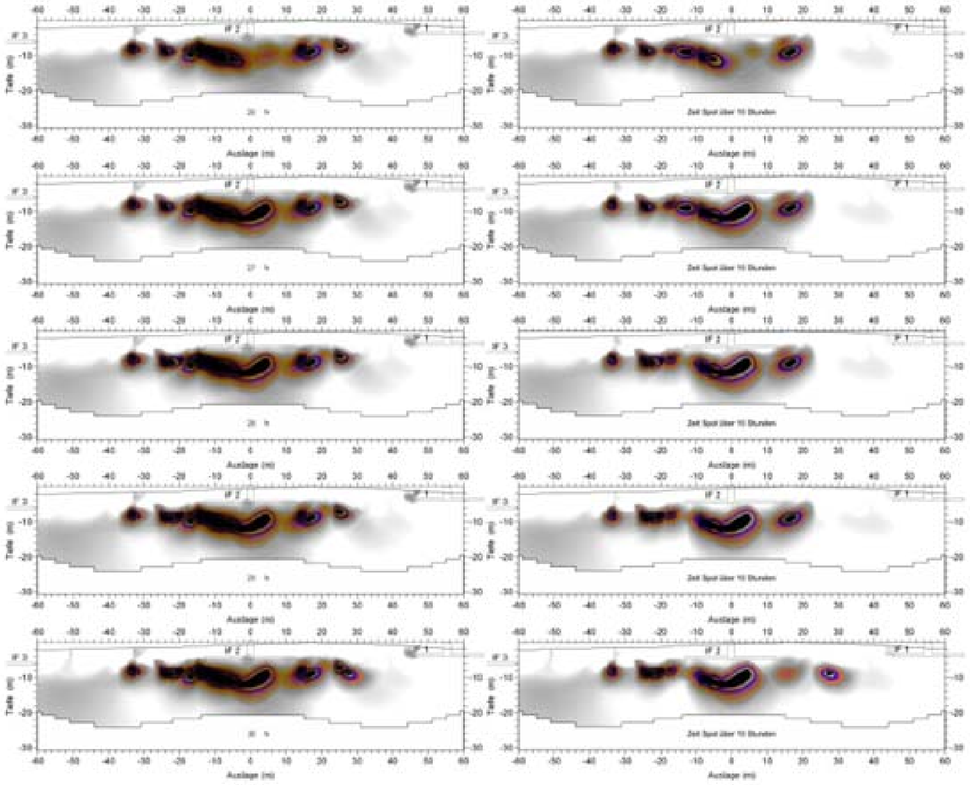

Figure 3.2 Time lapse image of a series of ERT images of an infiltration experiment. Right hand image is the situation after 10 hours at the same scale as the left hand image [30].

of the water flowed through 47% of the total water content. The analysis of breakthrough curves on a 0.14 m^3 waste sample [53] showed that 19 to 41 % of the total water content participated in the solute transport. In addition the amount of preferential flow was found to be dependent on the flow rate, in the sense that high flow rates enhance the preferential flow. Given the fact that landfills are unsaturated, the total landfill volume that participates in solute transport is relatively small.

In the first phase of the DFSL project, the extent of preferential flow was analysed in the pilot experiment with predominantly inorganic waste [91]. The emissions of mobile salts (Na, K, Cle) from a laboratory column leaching test were compared with the emissions at the pilot scaled. The results indicated that approximately 15 to 35 % of the total water present in the landfill body participated in the solute transport.

Geophysics and time lapse 3D measurement of water flow

Geophysical methods are used to visualize the transport of solutes within a landfill. Recently two PhD theses [18, 30] have been defended in which time lapse imaging of the apparent electrical conductivity with ERT has been used to obtain an image of water flowing through the landfill body. The results clearly show the effects of preferential flow; however the interpretation of these ERT data is still a matter of much (fundamental) research. Figure 3.2 gives an example of an interpreted 2D time-lapse image of an infiltration experiment. This example is an illustration of preferential flow resulting in a limited infiltration depth, possibly due to the presence of impermeable layers.

An important conclusion is that conceptual model of solute transport through landfills, formulated in the first phase of the DFSL project, still holds. The way solutes flush from the landfill is governed by mobile phases in contact with a large stagnant bulk, from which pollutants are only released by diffusion. Results from the international literature confirm this model and more and more research groups around the world seem to direct their research in the same direction.

Experimental evidence from these publications allow for some refinement of this conceptual model:

- Leaching behaviour in summer and in winter might differ significantly. In summer preferential channelling seems to be of much more importance than in winter;
- Once a preferential channel is established, certain mechanisms might make it a more stable situation. E.g. the waste becomes less water-repellent, thus increasing its hydrological permeability.
- Leachate, being the portion of the water content in the landfill draining from the landfill, flows through the preferential flow paths. Solutes from the immobile parts of the landfill exchange with the leachate via diffusion which is a very slow process. Generally, the presence of preferential flow paths will result in low concentrations in the leachate.

A draw-back of preferential flow is that it will also limit attempts to enhance biodegradation by leachate infiltration and recirculation because it will make it impossible for us to reach certain parts of the landfill. The concept of preferential flow has until now largely been ignored when designing systems for leachate infiltration. Understanding the hydrology of landfills will allow for the development of tools for monitoring and the development of full-scale technology for enhancing biodegradation.

3.1.1 Modeling of landfill hydrology

Modeling as a tool for predicting long term behaviour of landfills has already been used for a long time. The simplest non-physical concept for describing the water flow through a landfill is based on the so-called Liquid to Solid Ratio (L/S). The L/S is the weight ratio of the amount of water that has percolated through the landfill and the dry weight of the solids present in the landfill body. Sanchez & Kosson [55] give an example of a simple probabilistic approach for long term extrapolation of emission of chemical compounds. This L/S approach, however, does not take preferential flow into account.

Johnson et al. [26] present 4 modeling approaches in order to describe the Lostorf hydrological data. The comparison between the different approaches clearly showed that functional modeling approaches (empirical approaches based on a neural network and a linear hydrological storage approach) were better able to describe the data than mechanistic models used in vadose zone research such as Hydrus 5 and MACRO. The conclusion is that preferential flow through landfill bodies is a highly complex and non-linear process for which mechanistic descriptions are still lacking.

White and co-workers [101-102] have developed the LDAT model for landfills which is based on the concept of a generic spatially distributed numerical model that has been developed to contain and link sub-models of landfill processes in order to simulate solid waste degradation and gas generation in landfills. The model includes the simulation of the transport of leachate and gases, and the consolidation of the solid waste. The structure of the model consists of linked discrete constant volume elements.

Ustohalova et al., of the University of Duisburg-Essen have developed a constitutive 2-dimensional model based on the macro-mechanical theory of porous media (TPM) to simulate the long-term temporal dynamics of landfills. This model couples landfill hydrology to biodegradation, heat production, gas production and mechanical deformation. However, the model does not take preferential flow in to account [60].

McDougall has developed a similar approach which can be downloaded from the Internet (http://sbe.napier.ac.uk/HBM/). This model is an approach to describe settlement, water flow and bio-degradation of waste [39].

A major problem with using these deterministic models is that they rely heavily on parameters describing certain hydraulic properties of the waste. How to obtain relevant upscaled parameters for the highly heterogeneous waste material is still a matter of scientific debate.

Rosqvist, Destouni and co-workers [34, 51-53] model measured breakthrough curves with a range of Lagrangian stochastic models. This approach is based on deterministic concepts but the central focal point of the modeling approach is a concept called the travel time of a water parcel in a landfill. The models are based on a probabilistic description of these life times. All models could give an adequate description of the measured data, however, the parameters required to describe the data for the model based on the advection dispersion equation and the mobile-immobile (dual porosity) model) were unrealistic. The bimodal travel time model did describe the data with realistic parameter values. The main idea behind the bimodal travel time model is that water reaching the drainage system consists of two fractions, (1) water that moves along preferential flow paths in the landfill and as a consequence requires a very short travel time and (2) water that moves through the bulk of the landfill (slow diffusion type of transport process). The first type of water is hardly influenced by the waste in the landfill whereas the second type of water is in equilibrium with the landfill. As was mentioned in the previous paragraph, water in the drainage consists mainly of the first type of water. Recently the Lagrangian Stochastic Advective Reaction (LaSAR) modeling approach was coupled to the geochemical speciation model PHREEQC [34]. This is a first step for coupling the LaSAR and PHREEQC modeling approaches for flow systems where local geochemical equilibrium can be applied as a first step toward the development of a more general equilibrium and kinetic reaction system representation. This approach has the potential to enable the quantitative modeling of coupled bio-degradation and solute transport in heterogeneous preferential flow systems such as landfills.

3.2 Biodegradation

3.2.1 General

When organic waste is landfilled, organic material in the waste will be biodegraded by micro-organisms. Solid organic material is decomposed to soluble material and subsequently to biogas (a mixture of methane and carbon dioxide). The degradation of organic material is described as a sequential process of hydrolysis of the solid organic materials (e.g. hemicellulose, cellulose) into larger soluble organic molecules, fermentation of organic acids and methanogenesis.

In reality, all phases described above will occur simultaneously. When conditions are favourable, part of the cellulose at the outskirt of a piece of wood will hydrolyse and a biofilm will develop, in which degradation products ultimately are transformed to biogas. At the same time, biodegradation of the outer parts will open up the inner part of this piece of wood for

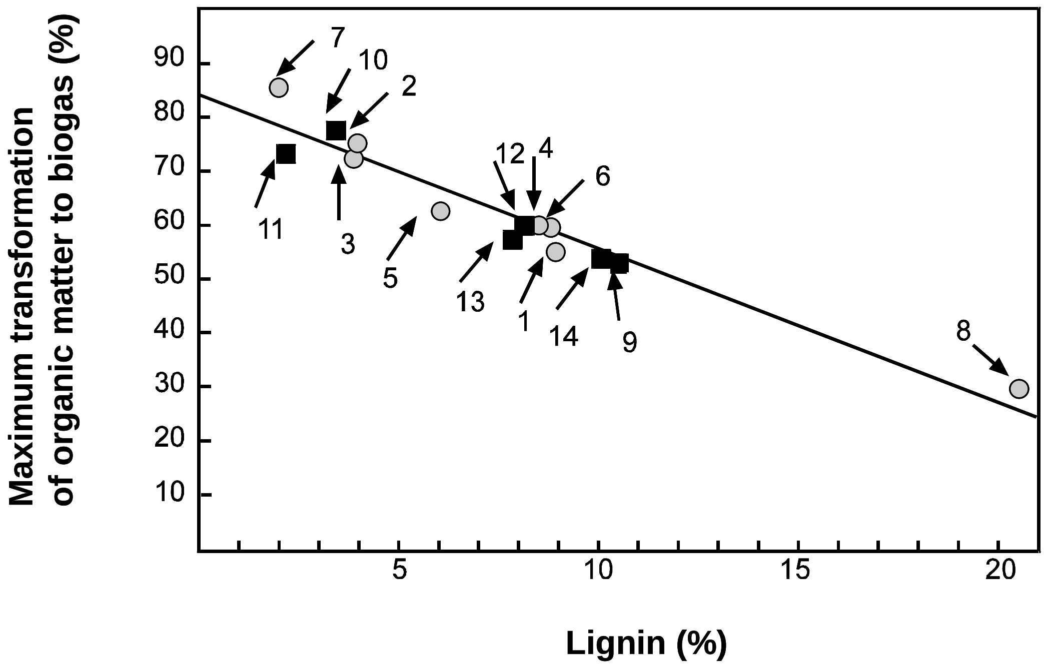

Figure 3.3 Relationship between fraction of organic waste ultimately converted and the lignin ration of the waste (1 = wheat straw, 2 = corn stalks, 3 = corn leaves, 4 = purple loosestrife, 5 = seaweed, 6 = water hyacint, 7 = corn flour, 8 = newspaper, 9 = elephant manure, 10 = chicken manure, 11 = pigs manure, 12 en 13 = cow dung) [10].

hydrolysis. So on the scale of a piece of waste biodegradation might also be envisaged as a reactive front, which slowly eats its way through solid biodegradable matter.

Not all organic material is biodegradable under anaerobic conditions, for example, lignin is very resistant against anaerobic biodegradation, and in natural setting it appears that lignin shields part of the cellulose from being degraded. So even when conditions are favorable, not all organic material is biodegradable. This is illustrated in figure 3.3, in which maximum anaerobic conversion of organic material is plotted as a function of lignin content.

Complete biological stabilization does not mean the complete biodegradation of all organic matter. Waste can be considered completely stabilized when all organic material is converted, that is biodegradable under anaerobic conditions.

On a larger scale, the heterogeneity of the waste is also a complicating factor. Due to differences in the environmental conditions (e.g. variation in water content, inhibiting conditions due to local chemical composition), biodegradation might be fast in some, slow or even completely inhibited in other spots. The biodegradation rate depends on aspects such as waste composition, waste management practices (homogenization, compacting, temporary and final lining) and local climate conditions (temperature and excess rainfall). The biodegradation in large parts of the waste is inhibited for longer times, and possibly permanently, unless the local environmental conditions change. Estimates for the amount of organic material in full-scale landfills that ultimately converts to biogas are obtained from landfill gas formation models. For municipal solid waste dominated by household waste in the Netherlands, the amount is estimated to be 58% of the organic material present [11]. However, it should be noted that these results are due to a combination of unfavorable

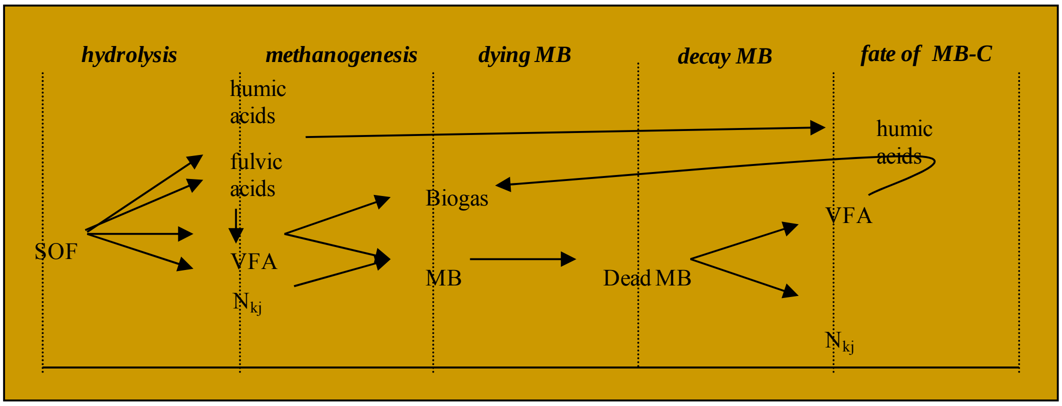

Figure 3.4 Degradation model as developed in the previous phase of the sustainable landfilling project.

conditions combined with limited degradability under anaerobic conditions. The aim of the sustainable landfilling approach is to improve the conditions so that degradation is maximized.

3.2.2 Modeling of biodegradation

There have been numerous attempts to model biodegradation of landfills. As with all models, an important distinction between different models is the application objective. On the hand, biodegradation models exist that predict overall biodegradation of the solid organic fraction and the resulting biogas production. These models generally are very simple, solely based on empirical data on biogas-production in actual landfills and extrapolation using first order (exponential) models, and therefore do not present further insight in mechanisms of biodegradation and e.g. impact of biodegradation on leachate composition.

On the other end of the application spectrum we find models based on complex deterministic approaches taking a multitude of reaction pathways in to account [22, 43, 47, 60, 101]. This type of models gives more insight in the processes and can predict leachate composition in detail and might be able to describe effects, such as inhibition due to VFA-accumulation or competition of methanogens with sulphide producing bacteria. However these models generally neglect variation in conditions present in a landfill that in reality might be the most important factor determining the overall rate of biodegradation. Examples are the heterogeneity on a micro-scale (not all solid organic fraction is available for hydrolysis from the start) or on a macro-scale where differences in waste composition, moisture content, nutrient availability are generally not described and possibly as the result of this, most deterministic models overestimate the rate of biodegradation [4].

One of the most important conclusions from the first phase of the sustainable landfilling project was that prediction of the amount and the quality of leachate released from a landfill requires the model to integrate biodegradation and subsequent production of degradation products, with the transport of these products through the landfill. It appears that transport is dominant rate-limiting step for virtually all realistic combinations biodegradation rate and flushing scenarios of the waste [86]. In order to adequately describe the leachate being generated at the bottom of a landfill, a model was used based on a simplified deterministic reaction mechanism where global kinetic parameters were estimated so that overall biogas production matches results from landfill gas formation models and which are in line with the values encountered in real landfills (see figure 3.4). The development in modeling landfill biodegradation of the past couple of years has not led us to change our conclusion. At the moment we feel that no further research effort is required in the Netherlands on further improving the modeling of landfill biodegradation and microbial processes. It will be sufficient to follow and keep involved with the progress worldwide, as many other groups are performing such research.

3.2.3 Nitrogen and biodegradation

One important exception to the conclusion above was the behaviour of nitrogen (N). Upon biodegradation of solid organic waste, nitrogen is released but immediately partially incorporated again in the microbiological biomass that is responsible for biodegradation of waste. This ultimately results in a delayed release of nitrogen, ultimately yielding nitrogen emissions with the leachate that might be increased for longer times (van Zomeren et al., 2006) [86]. In this phase of the Sustainable Landfilling project, no additional attention was paid to N-formation from waste.

3.2.4 Aerobic biodegradation due to air injection

Composting is a well-established aerobic treatment method of organic waste. The biodegradable portion of the Municipal Solid Waste is stabilized in a significantly shorter time frame (compared to anaerobic conditions). Injection of air in landfill bodies will result in a process that is similar to composting [46]. Organic matter will be degraded to humic like substances. Aerobic degradation will produce heat and as a result, temperature will rise in the landfill body. Literature has shown a large amount of very promising results, both on laboratory scale as well as large scale experiments ([8, 46]

Much more organic carbon will be degraded during aeration because compounds such as lignin which are non-biodegradable under anaerobic conditions can be biodegraded under aerobic conditions.

Aerobic conditions will result in an increase in redox potential which implies that sulfides will tend to oxidize, potentially causing heavy metals precipitated under anaerobic conditions to remobilize. However, in aeration projects carried out in Germany and at the Braambergen site, no evidence for remobilization was found. Our hypothesis is that aeration primarily occurs in the mobile zone (the preferential flow paths). Aeration will cause redox gradients to develop within the landfill body and our assumption is that biofilms will develop along these gradients. Heavy metals will be immobilized along the biogeochemical gradients. In addition, nitrification of ammonium and subsequent denitrification can occur in such biofilms, thus potentially causing a strong reduction of nitrogen in the produced leachate and methane will be oxidized by these biofilms as well, reducing the methane emissions. The humic organic residue remaining after the composting process, is stable, has a very large sorption capacity and as a result will absorb a wide range of micropollutants (such as the heavy metals), causing an improvement in produced leachate quality.

3.3 Inorganic geochemistry

3.3.1 Basic chemical processes

In this chapter, an overview of chemical processes controlling the release of contaminants from waste materials is given. It may seem surprising at first, but the total composition (in the sense of mg of an element / kg of material) has a limited influence on the maximum leaching of most elements. The release is the result of a number of geochemical mechanisms and physical factors such as dissolution, adsorption and redox reactions. The result of all these processes is that the leached amounts seldom correlate with the total amount present. An exception has to be made for the non-reactive and soluble salts such as ordinary kitchen salt

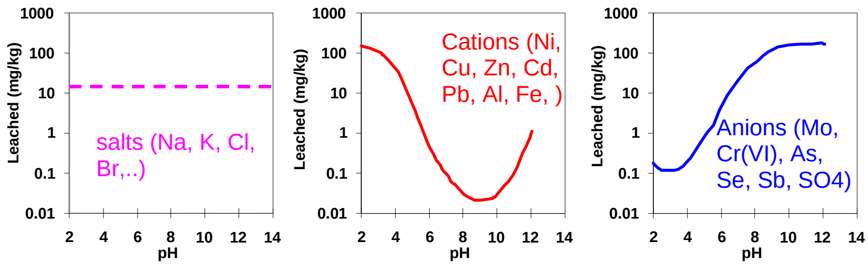

Figure 3.5 General leaching behavior of three groups of constituents as a function of pH. Cations, anions and soluble salts have a distinct leach pattern, caused by their chemical speciation, and vary orders of magnitude as a function of pH.

(sodium chloride). Here the maximum amount leached over time will be similar to the total amount present in the material [64].

Basic chemical mechanisms

Three different chemical mechanisms control the release of contaminants; the dissolution of a mineral (**solubility control**), adsorption processes (**sorption control**) or the total content in the waste material (**availability**). Some contaminants show affinity for adsorption to reactive surfaces.

pH

The release of many constituents in a wide range of materials (waste, sediment, cements, granular and monolithic materials,…) is strongly influenced by the pH of the material itself and the pH of the surrounding environment. The pH value of the surrounding fluid is a limiting condition which determines the maximum water phase concentration. Using this idea, pH-dependent release curves may be measured for each material. Release curves show similar and systematic behavior for different groups of elements, only the absolute levels may differ between different materials. Typical release curves as a function of the pH for salts, cat ions and anions are presented in figure 3.5. The similar behavior of release as a function of pH for different materials implies that the solubility controlling phases are the same in the different materials. The difference in absolute level from one material to another indicates differences in other (secondary) factors such as the amount of oxides (of iron, manganese and aluminium), clay and organic matter present. The strong influence of pH on release is explained by the fact that both dissolution of most minerals and sorption processes are highly pH dependent.

The natural pH values of materials may vary greatly. For instance, cement-based materials can buffer the pH of the environment to values around 12 (or higher), whereas predominantly inorganic waste buffers the pH to values around neutral (pH 6-8). The actual pH at which leaching takes place, depends on the pH of the material itself, the pH of the surrounding environment and the buffering capacity of the material. The general effect of changing pH values as a result of imposed environmental conditions is illustrated in figure 3.6. The total amount of a contaminant does not change as a function of the pH. Generally, the potentially leachable amount is also significantly lower than the total amount.

The potentially leachable amount is used as an input parameter in geochemical model calculations to predict the leaching behavior of waste materials. The red line shows the actual leaching behavior of metals (in this example Cd) as a function of pH. In a landfill, the pH of the leachate is around 6-8, indicated by the pink dashed box. The actual leaching behavior of a metal can change when the composition of the waste changes (i.e. more iron-oxides or clay

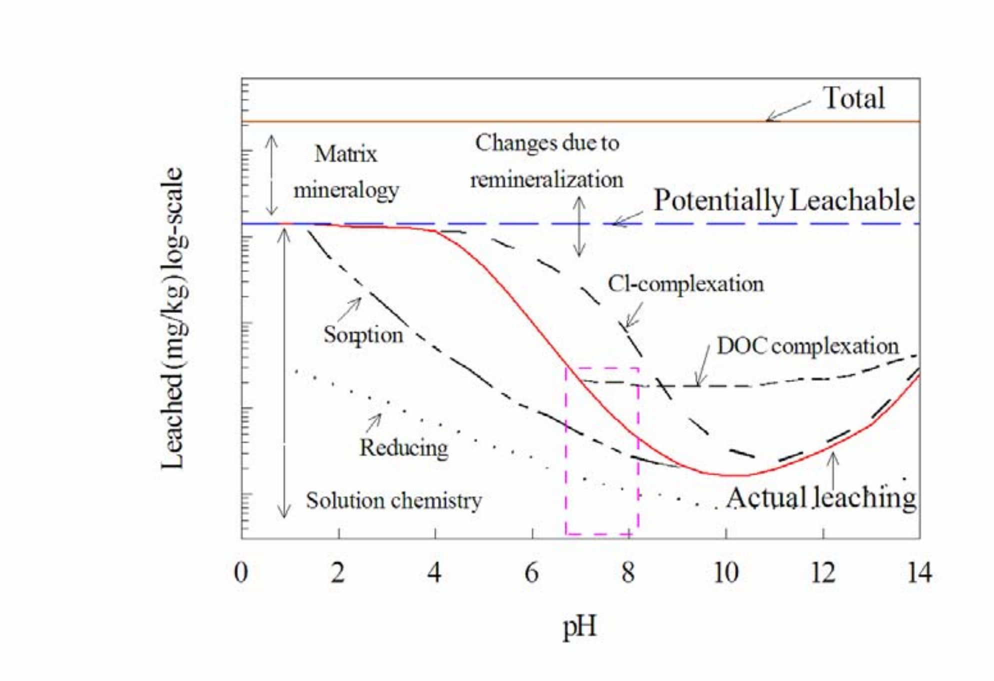

Figure 3.6 Leaching behavior as a function of pH versus total composition. In the figure, also the difference between the total compositions in the material is shown versus "potentially leachable" and "actually leachable" (the red curve) is shown. Note the log scale on the y-axis.

for sorption processes, more DOC causes increased leaching at neutral to high pH, a reducing environment generally causes a lower metal solubility). More information on specific chemical properties is given in the paragraphs below.

When the chemical processes that lead to release of contaminants are understood, a basis is formed for long-term prediction of the emissions from waste materials. The basic characterization and geochemical modeling approach is therefore an important part of the total environmental risk assessment of landfills. This approach was also followed in the first phase of the Dutch sustainable landfill project [63, 86, 90, 93, 98, 103].

3.3.2 Chemical form of the constituent in the waste material

Aside from these basic chemical mechanisms, the characteristic leaching behavior (e.g., the pH dependence shown in the above figures) of an element is determined by the chemical *form* of a contaminant. Contaminants may be present in the oxidized or reduced form (e.g., Chromium may be present as CrO_4^{2-} or Cr^{3+}). Because the solubility of these forms differs, it is obvious that the leaching behavior will depend on this form. The chemical form of elements is also called “speciation”.

Another example of speciation is the association of certain elements with organic matter. Heavy metals tend to complex strongly with natural humic substances present in natural waters, soils and waste materials. These organic complexed heavy metals are generally highly soluble and therefore, are released more rapidly than other forms of heavy metals (see also 'organic matter'). In the following paragraphs the most important factors controlling speciation will be discussed.

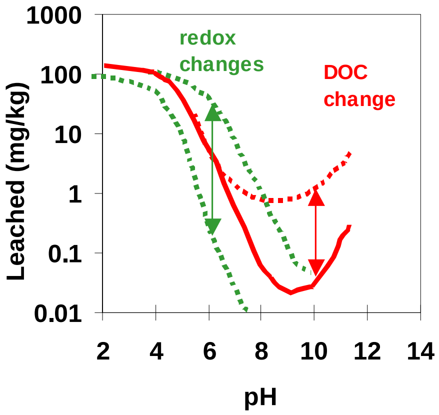

Figure 3.7 Absolute levels of leached amounts are different for each material due to influence of redox, DOC (dissolved organic carbon) and other factors. The leaching patterns of different groups of elements for all sorts of materials are very systematic, but differ in absolute levels (leading to a "chemical fingerprint" of a material).

Redox

The oxidation / reduction state of the material or its environment ("redox") influences the chemical form (speciation) of a contaminant. For heavy metals, the oxidation of an initially reduced material usually enhances leached amounts while reduction will have the opposite effect. This relates to the chemical form of the elements of interest. An example of the effect of the redox state of materials is given figure 3.6 and figure 3.7.

Acid-base buffering

The acid- base buffering capacity of a material determines how the pH develops over time under influence of external factors. Examples are the neutralization of cement-like products due to the uptake of atmospheric carbon dioxide. In such cases, the alkaline buffering capacity of the alkaline material determines the time needed until the pH drops from strongly alkaline (pH > 12) towards a neutral pH value (pH ~ 8).

Organic matter and DOC

Solid and dissolved organic matter or humic substances (often expressed as "DOC", dissolved organic carbon) consists of complex molecules that have a high affinity to bind heavy metals. The presence of DOC can enhance leaching by several orders of magnitude. As a result, a new partitioning between DOC-bound metal and free metal will be established. Organic matter is usually present in relatively large amounts in environments supporting living organisms (such as soils, sediments, sludges). An example of the effect of organic matter and DOC in waste materials is given in figure 3.6 and figure 3.7.

Composition of the water phase and ionic strength

The ionic strength influences the solubility of other components (generally, a higher salt strength increases the leaching of contaminants). Other components present in the solution may cause enhanced leaching due to complexation, such as metal complexes with chloride or carbonates.

Table 3.1 Summary of the main factors influencing release.

Chemical processes	Physical factors	*External factors*
- Dissolution	*- Percolation*	*- Amount of water,*
- pH	*- Diffusion*	*- Contact time*
- Chemical form	*- Surface wash off*	*- pH of environment*
-Total composition/ availability	*- Granular/monolithic*	*- Temperature*
- Redox.	*- Size (particles or monoliths)*	*- Redox of environment*
- Acid-base buffering	*- Porosity*	*- DOC / Adsorption*
- DOC	*- Permeability*	
- Composition water phase/ionic strength	*- Tortuosity*	
- Temperature	*- Erosion*	
- Time		

Temperature

Temperature increase generally leads to a higher solubility of contaminants. In addition, an increase in temperature has an increasing effect on chemical reaction rates, and thus also an increasing effect on transport by diffusion.

Time

Time is an important factor for the amount released when

- In general, the *time scale* applies to the disposal scenario of the landfill;
- The *rate* at which processes proceed, which may be limiting for the release in case of slow reaction kinetics (slow dissolution of minerals) or diffusion. It may not be feasible to allow such reactions to run to completion, as the time to reach that stage may be far too long. In that case, one has to estimate the possible consequences of such slow processes on the overall release.
- The change of material properties or environmental conditions over time. Examples are the degradation of organic matter (see also paragraph 3.2), changes in permeability of the landfill (see also paragraph 3), carbonation of alkaline waste materials (altering its release properties) or the increased surface area of a monolithic waste material due to erosion.

Test methods that include several steps provide insight in the short and long term effects of leaching. Such tests may give information for interpolation or extrapolation towards shorter or longer leaching periods. A summary of factors influencing release is given in table 3.1 (obviously, not all factors are equally relevant and depend on the scenario taken into account).

Example of geochemical modeling results based on a pH dependence leach test

The results of the pH dependence test and the results of organic matter fractionation and reactive Fe/Al-oxide extractions are used for geochemical modeling. With geochemical modeling, the underlying chemical processes leading to release of contaminants can be estimated. A general overview of chemical processes that can be identified in both the solid and the liquid phase is given in table 3.2.

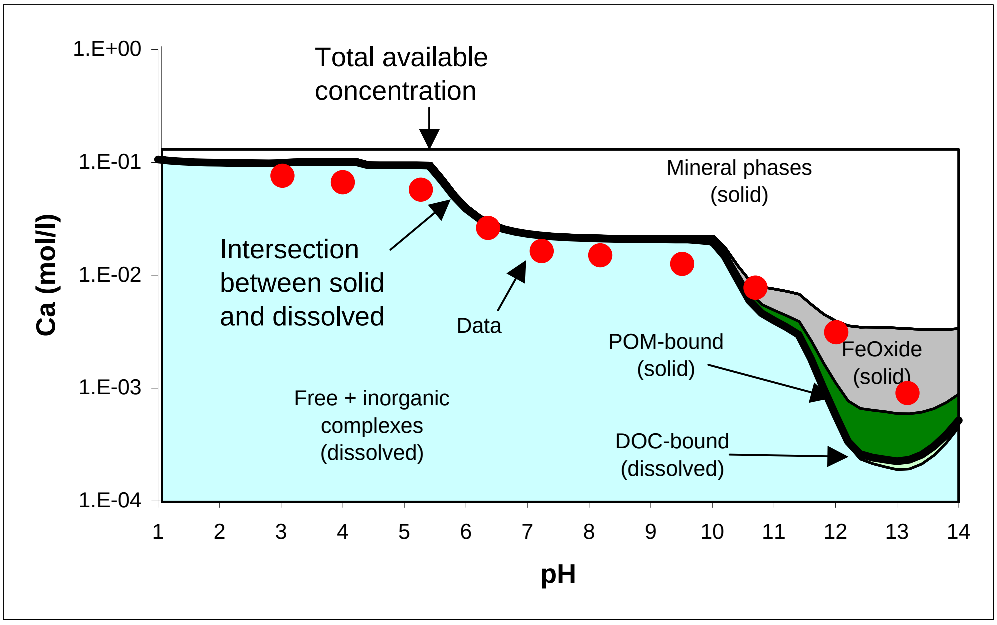

Figure 3.8 Example of integrated data presentation for pH-static leaching test results and geochemical speciation modeling. Red data points represent leaching data, black solid line is the predicted leached concentration. Areas represent the element speciation: White=minerals, Gray=FeOxide sorption, Dark green=complexation to solid organic carbon, Light green=complexation to dissolved organic carbon and Light blue=free+inorganically complexed form (van Zomeren et al., 2006).

An example of the measured and predicted leaching behavior is given in figure 3.8. The leaching data from a laboratory pH-static leaching test 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 for good understanding of the chemical processes that determine the leaching behavior. Moreover, the calculated chemical speciation of the element in both the solid matrix and the sample solution is also shown. The predicted leaching behavior is therefore the intersection (solid black line) between the calculated speciation in the solid matrix (minerals, sorption to Fe-Oxides and binding to solid organic matter) and in the solution (free + inorganic and complexed by dissolved organic carbon). This type of data presentation integrates the predicted total leached concentration as well as the different species that determine the leached concentrations. The upper line gives the total available concentration (input in model). The white area shows the amount of the element bound as minerals in the solid phase. Sorption to Fe-Oxides is represented by the gray area while complexation to solid organic matter is dark green. These sums of these areas represent the total amount in the solid matrix as a function of pH. In the leachate solution, the light blue area is the total amount of the free ion and the inorganically complexed form. The light green area represents the amount of the element that is organically

Table 3.2 *General speciation of contaminants in the solid phase and in the leachate of waste materials. The major phases and species are specified in both the solid phase as well as in the leachate.*

Solid phase	*Dissolved (leachate)*
Mineral phases (e.g. CuO, $Pb(OH)_2$)	*Free ion (e.g. Cu^{2+}, Pb^{2+})*
Bound to solid organic matter (humic substances)	*Inorganic complexes (e.g. $[Pb(OH)_4]^{2-}$)*
Adsorbed to Fe/Al-(hydr-)oxides	*Complexed to DOC (humic substances)*

complexed. The sum of the light blue and the light green concentration at a certain pH value is the predicted total dissolved concentration. Ideally, the solid black line crosses the data points at every pH value, implying that the chemical processes leading to release are fully understood using the geochemical model.

3.3.3 Application of an existing leaching test framework on waste materials

Release mechanisms and test protocols

Test protocols should provide the information from which in principle a mechanistic interpretation can be made. CEN TC 292 already developed such a testing framework, which will be discussed in this chapter. The basic characterization step in the test hierarchy (as specified in ENV 12920) should include the measurement of both major and minor elements, DOC and anions. Currently, the first step in the CEN TC 292 test hierarchy, are or will be:

- **Percolation test**, PrEN 14405 (up-flow percolation test to determine the leaching behaviour of **granular** waste materials under specified conditions). The test is performed using columns (30 x 5 cm) and the leaching is performed with demineralised water of natural pH (the material tested will superimpose its 'own' pH to the solution). Concentrations are measured in usually 7 different fractions up to a cumulative liquid to solid ratio of 10 L/kg (about 50 pore volumes). The choice of 10 L/kg is often representative for a long-term situation in practice. At the same time, results at L/S 10 make comparison with results of the pH dependence test possible (also performed at L/S 10). The test is designed such that local chemical and physical equilibrium is attained;
- **Tank test ('diffusion test')**: under development in CEN TC 292 with similarities to NEN 7375 (Determination of the Leaching of Inorganic Components from **Monolithic** Building and Waste Materials with the Diffusion Test) and other national standards (France, Austria and Nordic countries). A monolithic material is placed in a tank and is surrounded by water of natural pH. At specified times, concentrations in the leachate are measured and the leachate is refreshed. Total test time is 64 days and the results of the test are generally expressed in mg/m^2/64 days;
- **pH dependence test**, PrEN 14429 (Influence of pH on leaching with initial acid/base addition). The test is carried out on (crushed/grained) samples (<2mm) at a liquid/solid ratio of 10 L/kg, and various amounts of acid and base are added to obtain a final pH. After an equilibration period of 48 hours, concentrations of elements are measured in the filtered eluates. The test also gives an indication of the buffer capacity of the material. The test is designed to represent chemical and physical equilibrium conditions. A typical result of a pH dependence test under different circumstances is given in figure 3.6.

3.3.4 Basic data presentation and interpretation

For the *pH dependence test* and the *percolation test*, the results are preferably expressed in mg of substance leached per kg of dry solid matter (release units). The results can also be expressed as a concentration (mg/l) when necessary for specific aspects (e.g., evaluation of solubility control). The reason for this way of data presentation is that it enables comparison of results for different L/S ratios (the amount of water in contact with the material, expressed in L/kg) for groups of constituents showing similar release behavior (salts as well as solubility controlled release). Also, it makes a direct comparison possible between results from the percolation test and a pH dependence test.

It must be realized that, the concentrations in the eluate of a leaching test can generally not be related directly to the impact of soil, groundwater or surface water. The eluate from a leaching

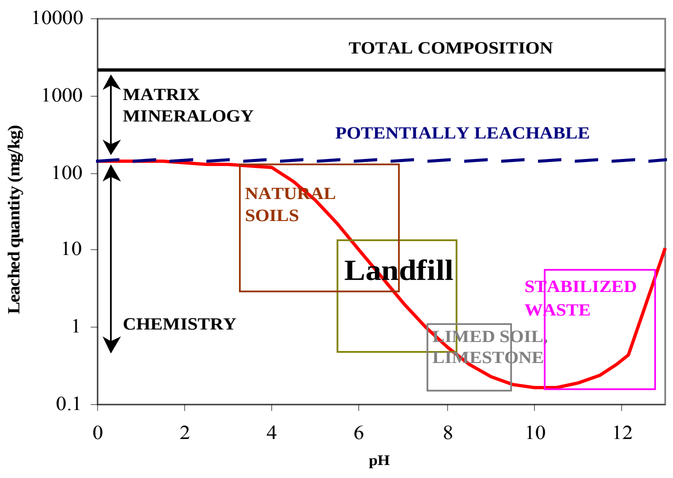

Figure 3.9 Standard presentation and interpretation of results from a pH dependence test (PrEN14429, expressed as release in mg/kg at L/S 10). The curved line illustrates a hypothetical release curve of a metal cation. The different pH values found in specific environments are indicated with boxes. Note that there may be a considerable difference between the total content of a material and the amount that is available for release (‘available’ = potentially leachable).

test reflects the release under the conditions imposed in the test. In an actual landfill situation, the first pore volumes (L/S=0,1-0,5) may have much higher concentrations, than the release after a longer period. Furthermore one should realize that many leached substances may be adsorbed first by the soil particles and may later be transported to the ground water. So the concentrations of substances in an eluate directly from a test will often be strongly different from the concentrations of substances in the landfill leachate after transport through the underlying natural soil layer before it enters groundwater. This mechanism is also graphically represented in figure 5.2.

The leaching pattern obtained with the *pH dependence test* is the result of a combination of the material-specific chemical factors that control release, such as the presence of organic matter and salts in the material, redox properties, buffering capacity (implicitly measured in the pH dependence test), ionic strength and chemical speciation. The test result also allows extrapolating the result to relevant field conditions - such as what happens with the release when the material is exposed to different environmental conditions (see figure 3.9). The result of a pH dependence test gives the amounts expected to be released under different exposure conditions. In addition, the test results form the basis for geochemical modeling of the dominant chemical processes leading to release of contaminants in the environment. These chemical processes can subsequently be used in long-term predictions of the mobility of contaminants in a specific disposal scenario.

Results from a *percolation* test for granular materials can either be expressed as concentration (mg/L) versus percolated amount of water (L/S ratio) or as cumulatively leached concentration (mg/kg). The latter is often preferred, because cumulatively leached concentrations (mg/kg) enables comparison with legislative values (Annex II of the Landfill Directive) and to compare emissions between samples. In addition, the released amount at L/S 10 can be directly compared to leached concentrations in the pH dependence test at L/S 10. Materials leach at their *own* (native) pH value in the percolation test.

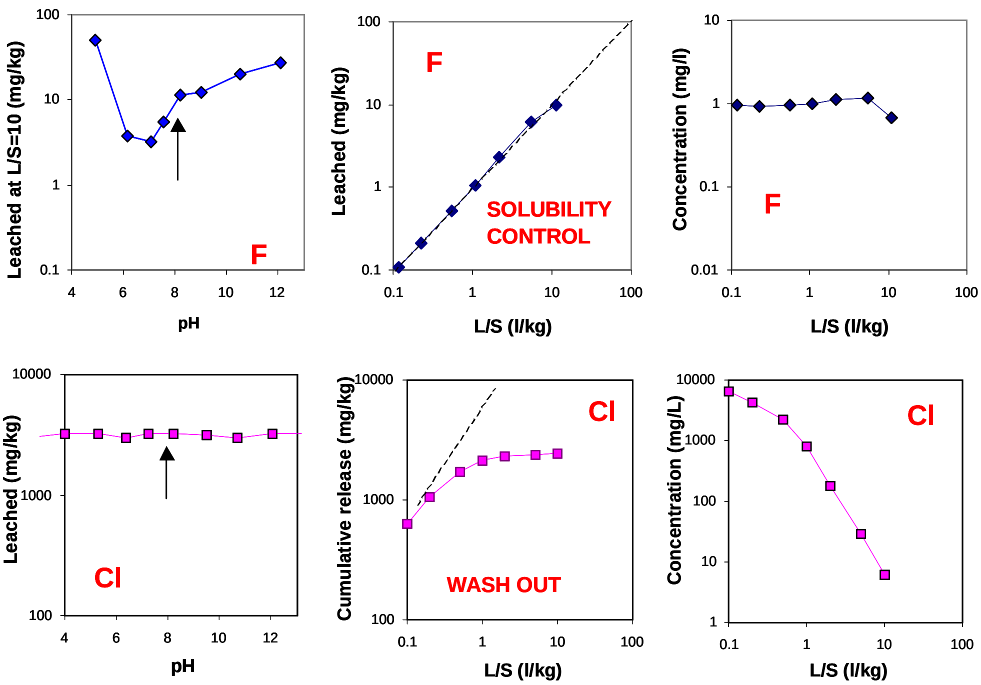

Figure 3.10 Data from a column leaching test on MSWI bottom ash. When a constant concentration is measured in each time interval (mg/L), this leads to a 1:1 slope in a cumulative leaching curve (case of fluoride). For soluble salts (e.g. Cl), that readily wash out, concentrations decrease rapidly as a function of L/S which leads to a cumulative slope lower than 1:1. The L/S scale (Liter water percolated/kg material) can be used for extrapolation of test results to field situations, relevant for impact assessment.

From the cumulative release curve, the underlying release mechanism can be identified for the entire range of L/S values, when the conditions during the test do not differ too much (e.g., pH) or for part of the range, when changes in major controlling factors occur during the test. The most important mechanisms include solubility control (dissolution of a mineral, e.g., $Pb(OH)_2$), and wash-out (relevant for non-reactive soluble salts such as Na and Cl). This is illustrated in figure 3.10.

3.4 Regulatory framework

The European Landfill Directive (LFD) is applicable to judge acceptance of waste materials at landfills. The LFD distinguishes three different types of landfills: for inert, non hazardous, and hazardous waste respectively. In the Annex 2 of the LFD a first step is made towards a source term definition controlling emission towards groundwater. The emissions of waste materials are to be measured using the percolation leaching test (PrEN 14405) and the results can be compared with the limit values for the acceptance of waste. For the sustainable landfill framework, the criteria of inert waste have been chosen as a target. It should be noted that the LFD is based on judgment of individual waste materials that are accepted (or rejected) on a landfill for inert waste. In the sustainable landfill project, the judgment is made on the total waste compartment after the active phase of process control has ended. This implies that a single waste material might not comply with the regulations but this is not harmful as long as the behavior of the total waste body is not unacceptably influenced.

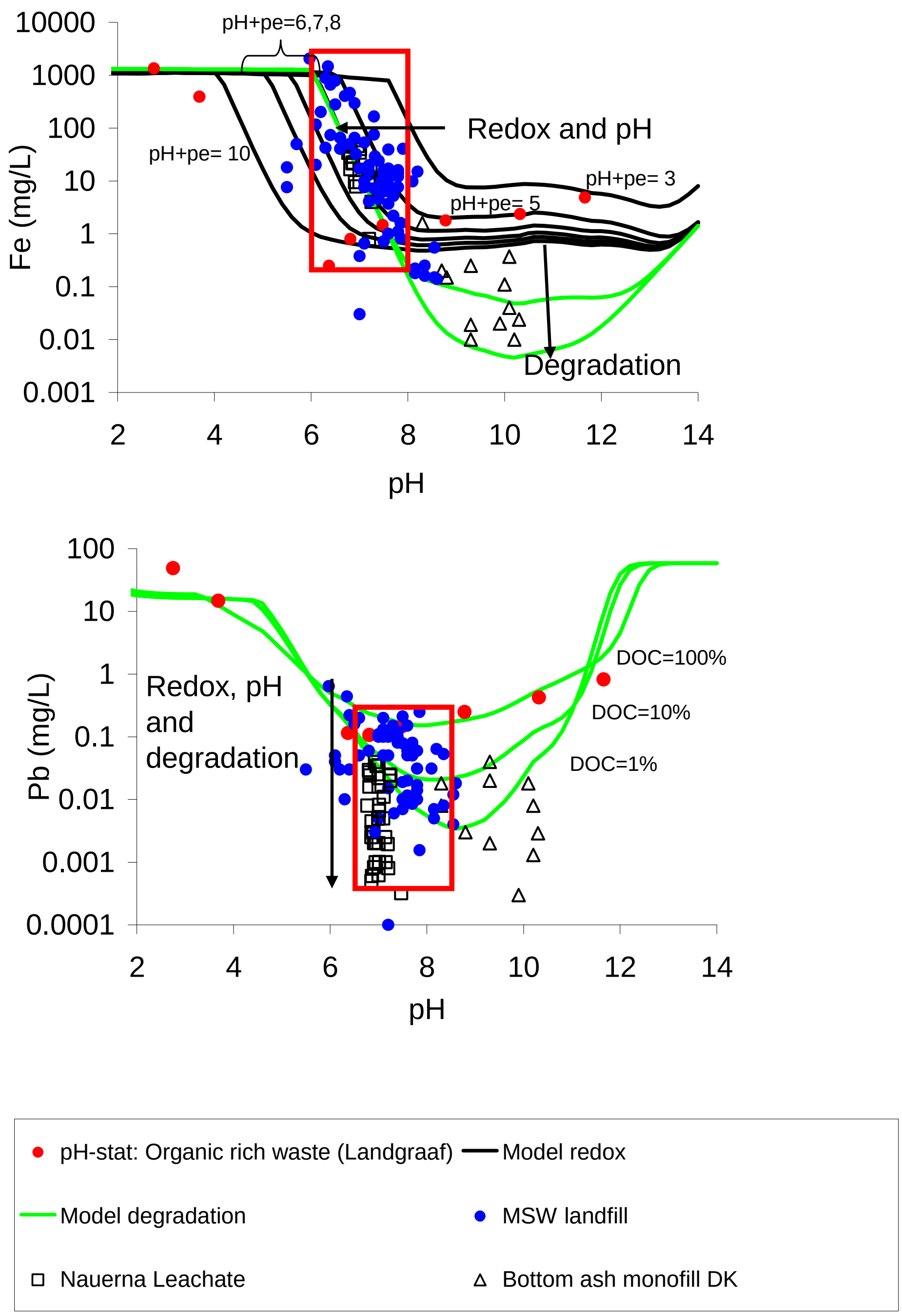

Figure 3.11 Results from geochemical speciation modeling of Fe and Pb for MSW. The leachate quality from MSW landfills, predominantly inorganic waste and from a MSWI bottom ash landfill are inserted for reference. Black solid lines are the model results from variations in redox and green lines represent variations in reactive DOC in the leachate.

The LFD has set criteria for As, Ba, Cd, Cr, Cu, Hg, Mo, Ni, Pb, Sb, Se, Zn, chloride, fluoride, sulphate, phenol index and DOC. In the sustainable landfill project, the emissions of bromide, nitrate, ammonium, Co, Sn and V are also taken into account to ensure acceptance of government and public. The WFD (Water Framework Directive) will put requirements on all activities affecting soil and groundwater (primarily groundwater). As such future emission levels from landfills will be derived from the WFD. This will inevitably introduce nitrogen as a relevant parameter.

3.4.1 Modeling of geochemistry

Geochemical modeling can enhance the knowledge on chemical processes leading to release of contaminants from soil and waste materials. The combination of leaching tests and geochemical modeling is a valuable tool in environmental risk assessment of contaminated soils, construction products, waste materials and landfills. The reader is referred to literature for an overview of the developed knowledge on relevant release controlling processes [2, 9, 12-13, 15, 23-24, 35-37, 41-42, 57-59, 62-63, 65, 67, 70, 74-76, 84-87, 96].

Monitoring of leachate concentrations from landfills is relatively straightforward and provides an estimate of the past and current release of contaminants. However, it has been found that leachate concentrations can vary substantially from one sampling round to another. In addition, monitoring only provides information on the current emissions and this does not necessarily imply that these concentrations will be stable on the long-term (e.g. due to degradation of organic matter or carbonation of alkaline materials). An important aspect in the emission of contaminants from landfills is the amount of DOC (especially humic- and fulvic acids) and the redox conditions in the waste body. The chemical speciation of elements can be influenced by variations in DOC and redox potential in the landfill. These changes are also likely to be largely responsible for variations in contaminant concentrations in leachate.

figure 3.11 shows an example of leachate data for iron and lead (various sources) as a function of pH. The red dots represent data from a laboratory leaching test on MSW where the pH was changed between 2 and 12 to study the pH dependent leaching behavior (pH-stat experiment). The blue data points are leachate concentrations from different MSW landfills in the Netherlands, UK and Denmark. The squares contain leachate concentrations from the Dutch sustainable landfill pilot experiment with predominantly inorganic waste (Equifill pilot study at landfill Nauerna). For comparison, leachate concentrations from a Danish MSWI bottom ash monofill were plotted as triangles.

The solid lines in the figure 3.11 represent the modelled solubility as a function of pH under different redox conditions (from pH + pe=3 to pH+pe=10). The solubility of iron is highly dependent of both the pH and the redox potential of the leachate. The pH-stat (laboratory) test is done under relatively oxidizing conditions (open to the air) and the modelled solubility of iron indicates that the system is oxidized (pH+pe=10). The leachate data from MSW landfills generally have a pH between 6 and 8 and are more reduced in comparison with the laboratory tests. The results show that the redox conditions (pH+pe) have to be in the range of 4 to 6, in order to explain the observed solubility of Fe. The expected iron concentrations in MSW landfills can then be narrowed to a limited range in pH and redox potentials. The red square in the graph indicates the system boundaries in terms of iron concentrations in MSW landfills. The results from modelled iron concentrations under conditions with more degraded organic matter show that this process does not influence the iron solubility in the pH range from 6 to 8. However, the effect of degradation on iron solubility is significant at pH from 8 to 12 (more relevant for alkaline waste materials and monolithic waste landfills).

A similar modeling study has been performed for lead. Preliminary calculations have shown that the solubility of lead is not dependent on the redox potential (data not shown). The modeling of the lead solubility as a function of pH was done for the assumption pH+pe=6, as was found reasonable from the modeling of iron solubility in landfills. The first calculation was done to describe the results from the pH dependence leaching test and this scenario was called “DOC=100%”. Next, the amount of reactive DOC (i.e. humic acids) was lowered to 10 % and 1% respectively. With the DOC decrease, also the total particulate organic matter is assumed to decrease to 75% and 50% of the original value (around 50 % is assumed to be residual). The graph clearly shows the sensitivity of the predicted lead solubility as a function of pH under these different conditions. The modeling results can also narrow the window of expected lead concentrations in landfill leachate. It can be seen that the apparent wide range in lead concentrations in MSW landfill leachate can be (at least partially) explained by differences in the amount of reactive organic carbon in the leachate. In addition, the results from the predominantly inorganic waste pilot experiment indicate that lead concentrations are indeed on the low side of the observed range of concentrations and they are also in agreement with degraded organic matter conditions. The red square indicates a range of expected lead concentrations between pH 6 and 8 for different amounts of reactive DOC in the landfill leachate. However, the effect of degradation on iron solubility is significant at pH from 8 to 12 which is more relevant for alkaline waste materials and monolithic waste landfills [66].

These examples show that geochemical modeling can help understanding the seemingly large and random variation in observed leachate concentrations. The results might indicate the system boundaries in terms of pH, redox conditions and organic matter content (also related to degradation of organic matter, see chapter 3. Future work should focus on data comparison for more landfill sites and application of the developed knowledge of geochemical processes on the extended dataset.

4. Landfill technology

4.1 Manipulation of landfill processes

From the moment it became obvious that landfills had an impact on the environment, technology and other approaches were developed to reduce this impact. Technology for emission control was considered from the start, the dominant focus was to reduce leachate production and emission by wrapping the landfill in low permeable geo-membranes. In previous research of the DFSL, options for process control have been identified and summarized [35, 37].

When implementing a new landfill all options for process control are available. However for stabilizing existing landfills, with waste already in place, possibilities for waste selection are non-existent. Solidification and solubility control is best performed by altering the initial waste composition as it is deposited so this can hardly be considered a realistic option for existing landfills. Therefore, the options are limited to enhanced biodegradation and flushing.

There are two major routes to enhance biodegradation; 1) Leachate injection and/or leachate recirculation results in an increased anaerobic conversion of solid organic matter to biogas. 2) Air injection results in an accelerated aerobic conversion of solid organic waste to carbon dioxide.

Since the early seventies of the last century, lots of experience has been obtained with both leachate infiltration and air injection. In Annex 6 of the generic report of the feasibility study for two pilot projects for emission reduction [79], an overview is given of 54 projects around the world. It should be noted that this overview is incomplete because the website http://www.bioreactor.org provides a list of many more bioreactor landfills in the USA alone. In the past decades this technology has been developed in a.o. USA, Japan, UK and Germany.

Table 4.1 *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 minimize 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;

About 10 projects have been carried out in the Netherlands with direct involvement of the members of the DFSL.

Leachate infiltration and aerobic landfilling both can be considered proven technology when we consider the fact that both techniques definitely lead to an increased rate of stabilization of the landfill body. Combined leachate infiltration and subsequent landfill aeration has shown (by monitoring the carbon levels in extracted landfill gas) that aeration can lead to a removal of up to 90 % of the degradable carbon present in the landfill before aeration (personal communication H. Scharff and K.U. Heijer). However, this still does not imply that these technologies are accepted as a means for emission reduction and alternative after-care. The reason for this is the lack of an accepted framework for assessing the emission potential of a landfill. Without such a framework it is impossible to assess the true benefit of many of the trials that have been carried out in the past as a means for reducing emission potential. Many of the trials were implemented in order to achieve different and less far-fetching objectives, resulting in different choices in technology, modes of operation and often incomplete monitoring. A number of trials have been carried out with different objectives:

- The earliest attempts to re-inject leachate were aimed at reducing the amount of leachate formed or to utilise in-situ biological processes in the waste to improve the leachate quality. In the 70's and 80's, this was common practice throughout the world. In the Netherlands experience was obtained with leachate infiltration in landfills at Bavel, Boeldershoek, Vlagheide, Haps en Zoetermeer [21]. Compared to leachate infiltration for enhancing biological stabilisation, these earlier projects infiltrated only small amounts of water and the monitoring was limited to the amount and quality of leachate generated;
- Since the beginning of the 1990's, efforts were undertaken in order to stimulate landfill gas production, increase the profitability of landfill gas recovery schemes, and minimize methane emissions after landfill gas recovery at a certain site became uneconomic. This was the main objective for the Dutch pilots in Wijster 1C and Elspeet, but also at the bioreactor demonstration at Yolo-county. Technology applied in these projects generally aim at stimulating biodegradation throughout the waste body by infiltration of large amounts of water in a homogeneous way. This technology might therefore be considered to be state-of the art. However monitoring of the results is often restricted to biogas production, and changes therein which are difficult to detect on a real scale (see chapter 0);
- The main objective of many bioreactor landfills and aerobic reactors in USA (http://www.bioreactor.org) is to enhance stabilization of the waste, increase headspace, and extend the life-time of a waste deposition site. These projects aim for a rapid degradation of the initial 60-70% of the biodegradable material (so rapid settlings and recovery of headspace), rather than complete biodegradation of the last 20-40% (determining the long-term pollution potential). Monitoring is often restricted to monitoring of settlement;
- Finally, a few projects have been carried out in order to reduce the pollution potential of the waste in order to reduce future risks of soil and groundwater pollution. This is the most far-fetching objective, combining the previous ones with additional prerequisites on quality of the stabilized waste. This was the objective of much of the UK-based work on landfill bioreactors, the bioreactor demonstrations in Wijster, Landgraaf and Vlagheide and all effects in this study.

4.1.1 Leachate infiltration current status – knowns and unknowns

Effect on biodegradation

At the moment it is widely accepted that leachate infiltration or recirculation enhances biological stabilization, landfill gas formation and accelerates landfill settlings. This effect is clearly seen at the bioreactor demonstrations in Wijster and Landgraaf. Abroad the project in Yolo County gives a striking example of a very effective accelerated biodegradation. These were all projects in demonstration cells, especially designed, constructed and filled for the infiltration project. So they are all based on fresh waste and relatively well monitored. Pilots to enhance biodegradation at existing landfills are much less conclusive. In paragraph 0, an example is given of the signal-noise ratio in landfill gas production at the infiltration in Wijster. This clearly shows the complexity of using measurements from biogas production systems to make conclusions about enhanced biodegradation. The experiences with leachate infiltration at Vlagheide (see chapter 5.3) are difficult to interpret as well after 4 months of infiltration.

Despite the large number of projects around, and the general consensus that leachate infiltration/recirculation at existing landfills significantly enhances the biodegradation of waste, the real effectiveness of application at full-scale is still unclear. A recent critical review of Benson et al. [6] of effectiveness of 5 bioreactors in USA supports this conclusion. They found little hard data that leachate infiltration actually was more effective in enhancing biodegradation than doing nothing. Barlaz a co-authors attributed this to inadequate design and operation (too little leachate infiltrated) and ambiguities in monitoring results. It can be expected that the conclusion of [6] will hold for the majority of leachate infiltration projects worldwide. Many projects have been inadequately monitored and the results of these projects are often poorly communicated.

Effect on waste quality

Leachate infiltration/recirculation is known to have a positive effect on waste-quality. Enhanced biodegradation results in a reduction of BOD, COD, N_{kj} and heavy metal concentrations in leachate. Infiltration causes an accelerated flushing of salts and other components which are normally not degraded. Both mechanisms have been convincingly proven at a lab-scale and have also been demonstrated to occur at the bioreactor demonstration in Wijster.

Mechanism - amount infiltrated

Full understanding of the mechanism behind enhancing the biodegradation by leachate infiltration is still lacking. Even though, this subject seems a bit academic, more knowledge and understanding of the process will have consequences for the way leachate infiltration is operated (and monitored). The hypothesis for the mechanism is that enhancement of biodegradation is the result of both increased moisture contents and increased movement of moisture. There is a lot of evidence for this hypothesis. To name a few:

- Klink and Ham [29] performed lab-scale-tests which indicate that moisture movement rather than moisture content determines biodegradation;
- In the bioreactor demonstration pilot in Wijster, the very wet parts of the waste were less biodegraded than moderately wet parts;
- In the experiment at Wijster 1C, enhanced gas formation was observed upon leachate recirculation, even though average moisture contents were not increased;

In chapter 3, a more in depth discussion is given on the processes responsible for enhancing biodegradation by leachate infiltration. As said above, the mechanism might be of importance for the way to operate leachate infiltration. It will not be enough to only raise the average moisture content of the waste, in stead a more sustained effort has to be maintained in order to stimulate movement of water throughout the waste. The amount of additional water to be added to the landfill is related to the optimal water content for biodegradation. In the Netherlands this amount is substantial compared to natural excess rainfall. Most Dutch projects aim for an infiltration/recirculation level of 1,500 mm y^{-1}, which is four to five times the net infiltration under 'normal' conditions.

Technology for infiltration

Various different technical approaches to leachate infiltration have been used: spray irrigation, deeper vertical wells, horizontal drains, shallow injection baskets, and injection fields. In the Dutch projects until now, horizontal drains have been the preferred technology, unless not feasible. In the Wijster, Landgraaf and Vlagheide pilots, horizontal drain systems were implemented. At Wijster 1C and at Elspeet an impermeable top-liner system was in place, so a number of small and shallow infiltration wells were made through the top-liner system. Little information was available on the distance between infiltration wells. Based on a theoretical calculation [45] distances of about 10 meters were considered acceptable.

For the feasibility analysis of the Kragge and Wieringermeer pilots, a review was made of infiltration techniques [79]. This review has been based on experiences and results of a great number of full-scale infiltration projects at landfills worldwide. Technical methods for water infiltration must be planned in such a way that water infiltration will occur in a controlled and homogeneous manner. This requires that preferential flow must be avoided as much as possible. However, preferential flow cannot be prevented completely. An evaluation shows that with current knowledge and experience, infiltration using horizontal trenches and infiltration fields are the best applicable methods. Shallow vertical wells (lances) spaced at short distance might be considered the best solution when an impermeable top-liner is already in place.

Infiltration via a permeable temporary surface cover might have a special position in this assessment as it is a rather "natural" infiltration method that should be obeyed as an additional effect on the water budget in parallel to one of the other methods. For the project in Kragge, a design has been made, based on the concept infiltration fields.

4.1.2 Technology for aeration knowns and unknowns

Aerobic stabilization is the second major route to enhance stabilization of landfills. The technology was first introduced in Japan as the Fukuoka method [38], found application at various landfills in USA [46] and is used in Europe, mainly in Germany and Austria [48, 50]. During aerobic stabilization air is injected in the landfill, causing the redox potential in the preferential zone to increase and as a result aerobic biodegradation will occur. The process is similar to composting. The increase in redox potential significantly increases the rate of degradation, rates of settlement etc. The idea is that that methane production will drop to nearly zero during aeration, although this still yet has to be proven by measurements. In most USA-based projects the aim is to recover headspace, with improved leachate quality as a side effect; whereas in European projects aim to achieve more complete biological stabilization of the waste and to reduce its pollution potential.

The current idea is that aerobic in situ stabilization is feasible when landfill gas production has dropped to low values (collection and treatment is still required but economic utilization of methane as an energy source is no longer possible). Consequently, aeration is a means to avoid long-term and cost-intensive treatment of gas of a poor quality. In addition aeration will improve leachate quality because the dissolved organic carbon concentrations will reach significantly lower values and reduced nitrogen will be oxidized and subsequently denitrification will occur transforming the nitrogen species to N_2, which is the major component of the earth's atmosphere.

Ritzkowski et al. [48], state that landfill aeration is also an attractive technology which can be applied to old landfills without a bottom or a top liner. Landfill aeration is a means to prevent unwanted methane emissions and leachate quality will improve, thus significantly reducing the risk to the environment. In comparison to leachate injection, much less detailed and quantitative information is available on aerobic landfills, despite the large number of field trials available. In appendix 6 of [79] an non-exhaustive overview is given of aerobic landfill tests.

Technical methods for aeration

Technology for aeration generally consists of vertical wells for air injection wells. In Europe air injection wells are often combined with wells for air extraction, and the injection – extraction is alternated. This is done in order to limit the possibility of unwanted emissions and to optimize the distribution of air in the landfill. It is also possible, however, to only inject air or only extract gas from the landfill. In the last case, ambient air will be pulled into and through the landfill body.

Aerobic conversion of waste is largely a microbiological process, with most reactions taking place in the water phase. Since aeration tends to dry out the waste (by increasing the temperature in the landfill body and removing a large amount of water as vapor via the gas phase), infiltration of additional water is often required to stimulate biodegradation.

Results from existing aeration projects allow us to derive some guidelines for design and operation of aeration pilots. The choice of the distances between wells is at the moment 25-30 m and seems to be a compromise between effectiveness and costs. The amount of air required for aerobic stabilization can be estimated from the amount of waste available and the estimated degree of stabilization. In existing aeration projects, monitoring of O_2 and CO_2 content of extracted air suggests that when well designed, about 60-75% of the oxygen injected can be consumed. As a result 100 m^3 of air per tonne of dry waste is assumed to be required for full stabilization of a landfill, on the condition that the spontaneous anaerobic stabilization is already almost completed.

Little to no information is found on the consequences of landfill heterogeneity on the completeness of the aerobic degradation. It is highly unlikely that the complete waste body will be aerobically degraded. Our hypothesis is that the air will migrate through the landfill body through the same preferential flow-paths as the leachate. Here most organic material will be degraded aerobically and the solid phase in the direct neighborhood of the preferential flow paths will oxidize as well. Depending on how long the air is injected, due to diffusion an oxidation front will slowly migrate further and further in to the immobile zone. As a result a redox gradient will develop at the boundary of the preferential flow paths. Micro-organisms will develop biofilms along these gradients creating niches for the degradation of a wide range or organic compounds including micro pollutants such as per-chloro-ethylene (degraded in the highly reduced zones) and compounds such as benzene and monochlorobenzene (degraded in the aerobic zones). The redox gradients are extremely beneficial for degrading these micro pollutants as benzene will not degrade under anaerobic conditions and perchloroethylene will not degrade under aerobic conditions.

4.2 Control of landfill risk

4.2.1 Application of monitoring techniques

In contrast with the technology for leachate injection and air infiltration, technology and concepts for monitoring, giving information on actual processes in the landfill and allowing for long-term quantitative prediction of leachate quality are underdeveloped. Development of monitoring approaches and the interpretation of the results has been one of the important aspects of the project. Monitoring is important for a number of reasons:

- It will allow the risk potential or the expected pollution load of an existing landfill to be quantified, based on more mechanistic insights (e.g. based on degree of stabilization of the waste, existence of larger pockets that are lagging behind in stabilization, resulting pH and redox in the waste) ;
- It will allow a more in depth assessment of causes of elevated risk potentials due to landfill heterogeneity and less optimal stabilization methods: how is the emission potential of the waste distributed throughout the landfill;
- Design and implementation of stabilization technology can be optimized.

The problem we face is that no single measurement technique available, will meet all objectives described above, so different approaches have to be combined. In the recent years two approaches have been followed in two projects carried out in the Netherlands, both co-funded by the DFSL. In the first, TNO and Deltares have been working on a risk assessment toolbox, based on process monitoring in the waste. For this purpose a number of monitoring techniques have been evaluated, including a range of geophysical techniques. At the moment, this type of monitoring will only lead to a qualitative risk-assessment.

The second approach was developed in the feasibility study [79], an approach based on ‘key performance indicators’ (KPI). The KPIs should enable the quantification of the success of technological measures applied to reduce the emission potential of the landfill. This approach is based on combining a number of indicators, which by themselves cannot be conclusive, but when considered together and in mutual interdependence provides a more meaningful interpretation. This concept will be further developed in the two feasibility projects.

Both approaches are based on the assumption that both the degree of biological stabilization of the waste and the hydrology of the landfill are important factors for assessing landfill stabilization. Methods to estimate the initial amount of degradable organic carbon (DegOC) landfilled and the amount of DegOC being converted to landfill gas are used in the well-known landfill gas formation models [56]. The accuracy, however, of these models is limited since the original amount and composition of the waste landfilled is often not well documented and the models are in principle empirical, resulting in large modeling inaccuracies. Fact however is that this type of mass balance in organic carbon is at the basis of most designs for accelerated stabilization of waste. Consequently the traditional landfill gas models have played an important role for the design of the new pilots at Kragge II and Wieringermeer (e.g. with respect the choice for leachate infiltration or aerobic stabilization).

Determination of degree of biological stabilization

There are various ways to quantify the degree of biological stabilization. Table 5.2 gives an overview of a number of a few possibilities. Nearly all approaches are based on the concept of the carbon mass-balance. The actual amount of DegOC in the waste is estimated from the amount of degradable organic carbon at the moment of landfilling ($DegOC_{deposited}$), minus the amount of landfill gas formed in the course of time ($DegOC_{emitted}$ + $DegOC_{extracted}$).

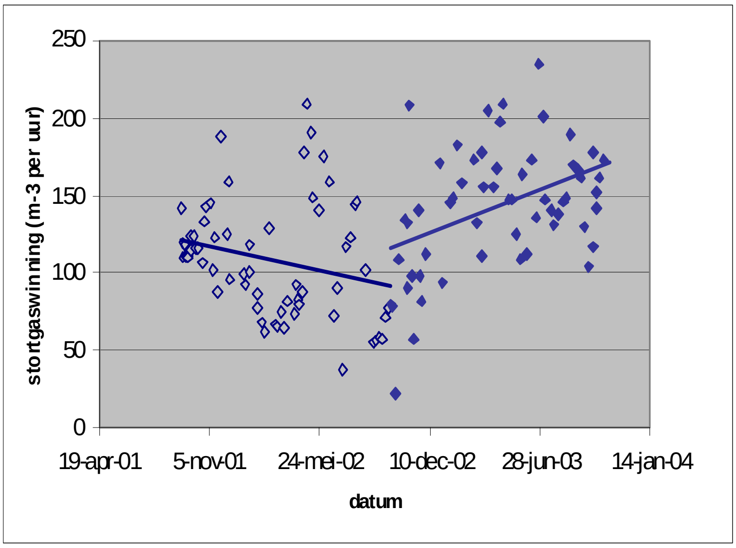

Figure 4.1 Development of landfill gas recovery before (open markers) and after (closed markers) start of infiltration at the Wijster project 1C.

$DegOC_{actual} = DegOC_{deposited} – (DegOC_{extracted} + DegOC_{emitted})$

Irrigation and recirculation of leachate will stimulate gas production and often more gas will be extracted as well. However, experiences at Wijster 1C, Elspeet and Vlagheide, test 3, indicate that interpreting the measured results of amount of gas produced and extracted is difficult and large uncertainties have to be accounted for. There are a number of reasons for this:

- The system of biogas extraction is often not designed for monitoring the test. Especially, when the test-cell is not sealed off from the surrounding waste it might be difficult to relate gas wells to the infiltration. Generally only under-pressure and gas composition is measured. The amount of biogas formed is generally only quantified upon gas utilization, and this includes gas from other cells as well;

Biogas production

Biogas production is often a difficult parameter to estimate due to two important facts:

- The background gas production and extraction levels are not known very well prior to leachate infiltration because gas formation is monitored at low frequency and often not well documented;
- The start of an infiltration project draws the attention of the landfill owner to biogas extraction, resulting in a better maintained system, which in itself can be a reason for increased biogas recovery.

Figure 4.1 is illustrative for the landfill gas production rates in an infiltration project. Clearly quantification of the additional production related to the infiltration is difficult to identify (the lines are fitted by the spreadsheet program) and in order to achieve a significant result, intensive monitoring is required.

Waste sampling and analysis for remaining potential biodegradation

The extent of biodegradation can also be determined from samples taken from the waste and subsequent analysis. A key problem is how to take representative samples from waste and subsequently reduce the sample to a size that is practical for analysis. Procedures for sampling and methods to determine how many samples of which size are required for significant conclusions need to be developed. Once a sample has been taken, several analytical procedures are available:

- Loss on ignition of waste samples is primarily linked to organic material present in the waste. It is one of the easiest ways to get an indication of the degree of stabilization, but it is not an accurate indicator. Large amounts of organic carbon (plastics, leather-like material, and material which is rich in lignin) can be stable under aerobic and/or anaerobic conditions. Loss on ignition always results in an overestimation of remaining biodegradable

Table 4.2 *Possible indicators of biological stabilization*

	Method	*Advantages/disadvantages*
Overall C-mass balance; gas extraction	Based on estimates of amount and composition of waste in the landfill combined with measured (and modeled) methane and carbon dioxide gas formed in and emitted from the landfill. Successful infiltration or aeration should result in significant changes in landfill gas composition.	*Results give information on the scale of the complete landfill.* *However, results are uncertain due to unknowns in waste amount and composition, leaks in monitoring system and uncertain model parameters. Applicability is reduced when test-cell is part of a larger system and gas extraction is not separately monitored or when history of gas extraction is less well known. Monitoring gas production is not a good indicator as landfill operators tend to optimize landfill gas extraction so that* CH_4 */*CO_2 *ratio is optimal. This severely reduces the value for monitoring.*
Ignition loss	Measurement of water and total C in waste samples. Possibly combined with manual separation of plastics. Gives both degradable and non-degradable C (plastics, leather, lignin-fraction)	*On a per sample basis relatively cheap (to be performed on 100 g to 1 kg scale). However, given the expected heterogeneity of the landfill an unknown (but probably large) number of samples will be required to obtain a representative result.*
Elemental analysis	Complete oxidation of waste sample, and measuring CO_2-formation. Gives both degradable and non-degradable C (plastics, leather, lignin-fraction) and possibly also inorganic carbon (carbonates)	*On a per sample basis relatively cheap (to be performed on 100 g to 1 kg scale). However, given the expected heterogeneity of the landfill an unknown (but probably large) number of samples will be required to obtain a representative result.*
Fibre-analysis	Speciation of organic material in soluble components, hemicelluloses, cellulose, lignin	*Gives detailed insight in composition. Expensive and performed at very small scale (< 1 g) and therefore with large sampling errors*
DOC speciation	Fractionation of DOC in terms of humic-, fulvic- and hydrophilic acids[83]	Quick and relatively cheap method for assessment of easy degradable (hydrophilic acids) and more recalcitrant (HA, FA) organic matter. The method does not assess the stability of the organic matter but gives a approximation for the stability of organic matter and the complexing capacity of DOC.
Fermentation test	Anaerobic fermentation of a sample in a batch reactor, and measuring volume of biogas generated during a certain test period (typically 21-72 days)	*Relative accurate indicator of remaining potential of under anaerobic conditions biological degradable material. Possibly performed on a scale of 0,5-1 kg; however most tests are performed at a smaller scale. Relatively expensive and slow.*
Respiration test	*Batch tests, during which biological conversion of the test material is measured from oxygen consumption during a test-period (typically 4-7 days)*	*Accurate indicator of remaining potential of degradable material under aerobic conditions. Accuracy as an indicator for anaerobic potential is questionable. Possibly performed on a scale of 0,5-1 kg; however most tests are performed at a smaller scale. Relatively cheap and fast, but to get representative results lots of samples are required.*

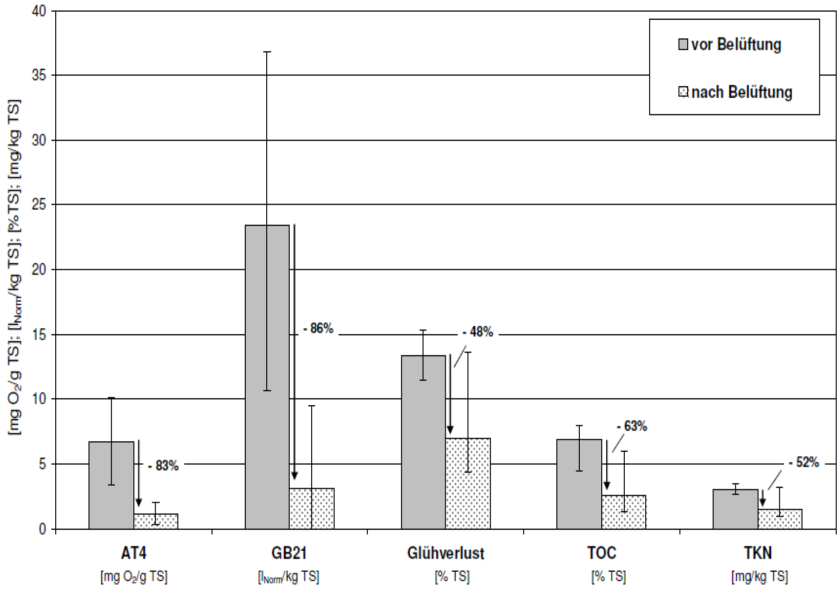

Figure 4.2 Results of different techniques to quantify the amount of stabilization based on 70 samples before and after landfill aeration [49]

organic matter;

- Oxygen consumption of the waste is determined during a certain period of time in respiration tests. Often used parameters to compare types of waste are AT4 and AT7 (Atmungsaktivität – oxygen consumption activity - during 4 and 7 days) or RI4 or RI7 (respiration index during 4 or 7 days);
- Cumulative gas production is measured in (anaerobic) fermentation tests. One method is the GB21 (Gasbildung - gas formation - during 21 days). However, research reveals that this period is too short to obtain a reliable indication of gas formation potential. In this period only 5 to 60% of total potential is being formed [7]. Another method (UK, France) is BMP (biological methane potential) which lasts about 100 days (personal communication H. Scharff);
- The total organic carbon content is measured by first removing all inorganic carbon by acidification and aeration and then subsequently heating the sample to temperatures in excess of 800 °C, where all organic matter is burnt. The measurement is based on the cumulative amount of CO_2 –produced during burning;
- The organic matter present in the waste can be characterized by a number of extractions under various conditions, ultimately yielding the amount of simple organic components (fats, sugars), hemicellulose, cellulose and lignin.

A number of the tests described above were tested at the aerobic landfill demonstration at Kuhstedt [49]. Figure 4.2 gives the overall results, based on 70 samples both prior and after aeration. For monitoring progress of biodegradation all indicators seem to be applicable, though the relative change in value is highest for the respiration test and the fermentation test, which is not surprising because these to methods mimic the natural degradation processes occurring in the landfill body.

However for determining the remaining potential of biologically degradable material only fermentation tests or respiration tests seem to be applicable. Due to a significant difference in costs it can be interesting to determine remaining potential for biodegradation under anaerobic conditions in a respiration test. Materials that degrade under aerobic conditions are to a large part the same as materials that degrade under anaerobic conditions. However it is also likely that under aerobic tests slightly more material degrades and therefore a slight overestimation

of biodegradability might be obtained, that becomes significant when waste is almost fully stabilized under anaerobic conditions. However when choosing a method one should bear in mind that fermentation tests themselves might not be accurate as well, since the experiment has to be cut off after some time. An additional consideration might be that anaerobic degradation alone may turn out to be insufficient to meet stability criteria. If an aerobic 'post-treatment' is necessary, it is very appropriate to apply a respiration test.
Based on previous experiences overall C-mass balance, ignition loss and fermentation tests or respiration tests are recommended for future pilot projects.

4.2.2 Determining moisture content and distribution

Water content and water movement is the second parameter that is of importance for assessing landfill risks. At one hand water affects biodegradation. Most biological processes

Table 4.3 *Possible indicators of moisture distribution/hydrology*

	Method	*Advantages/disadvantages*
Mass-balance	Measuring in- and outgoing water, both on a short term and on a long term	*Simple way to check on changes in average moisture content. Short-term changes give an impression of exiting short-cuts. Accuracy of the method depends on the support scale of the measurement.*
In-waste moisture sampling	Measuring moisture content, using probes. Many possibilities exist, often based on conductivity or impedance of moisture around probe.	*Continuous measurement of changes in moisture content. Combinations with temperature possible. However only waste in direct contact or in direct vicinity of the sensor. So many probes required for view on heterogeneity of moisture distribution. Often cross-correlation with leachate quality.*
EM	External geophysical measurement method with appropriate tool. Interpretation of results gives average conductivity of the top 5 meters of waste. Conductivity is determined by a.o. moisture content and moisture composition.	*Relatively cheap method, compared to e.g. geo-electrical sounding. However gives only average conductivities of a relative shallow layer.*
Ground-penetrating radar	External geophysical measurement method with appropriate tool. Method identifies objects below surface	*Capable to detect objects (water-lenses, plastic tubes, metal vessels), but only applicable for the first 2-3 meters*
Geo-electrical sounding	External geophysical measurement method with appropriate tool. Interpretation with reverse modeling results in a 2D or 3D distribution of resistivity in the waste.	*Most likely capable to give qualitative information on moisture distribution in the waste (unsaturated zones, saturated mobile, saturated stagnant). Possibly also applicable to identify effect of leachate injection from a well. Expensive method, with limited accuracy especially at deeper parts in the waste*
Seismic surveys	External geophysical measurement method with appropriate tool. Interpretation of results gives an indication of distribution of mechanical properties (stiffness) of waste.	*Insensitive method, unable to distinguish between wetter and dryer waste*
Tracer tests	*Adding liquid to the waste with a known concentration of a component, normally not occurring in leachate. Measuring and interpreting its release.*	*Gives quantitative information on mobile zones and mass-transfer from/into the stagnant bulk. Complicated and time consuming method. Application using lower drainage system only still not proven.*

occur in the water phase, so when waste is dry, biological stabilization generally is slow. Water movement is also important for keeping conditions favorable for biodegradation, e.g. by removing reaction products that inhibit further biodegradation. There are a number of ways to get an impression of moisture distribution and movement in the landfill. The most important ones are listed in table 4.3.

Water mass-balance

A water mass-balance is a relative easy way to assess changes in average water content in the waste. It is of special interest when infiltrating or recirculating leachate to enhance biodegradation. Accumulation is calculated from water/leachate addition, leachate extraction, excess rainfall, runoff and water vapor extracted with biogas. In this case not only a long term balance can be interesting, but also the reaction of the system to starts and stops or e.g. pulses of water.

In-waste sampling

One method to determine moisture distribution in the waste, and possible changes when infiltrating leachate is using moisture sensors in the waste. Various moisture sensors are available, many of them developed for agricultural purposes. In the bioreactor-demonstration at Wijster, 32 combined moisture/temperature sensors were dug-in. The sensors consisted of a block porous material that sucked in more or less moisture, dependent on the humidity of the waste. In this block, the conductivity was measured. The conductivity however depends on both moisture content and moisture quality and since quality changes in the waste, the signal only provided a more qualitative indication of

Geophysical methods

Geophysical monitoring techniques are well known from characterization of subsoil. These techniques generally do not directly measure moisture content, but generate information or identify heterogeneity that helps to understand moisture distribution in a landfill. In the past few years a number of geophysical techniques were tested at Dutch landfills (see chapter TNO-CoFi).

- EM-measurements use an electromagnetic signal that is sent and received by one apparatus. In this way an average conductivity is obtained from the subsoil to a maximum depth of 3 to 6 m. At Vlagheide electromagnetic conductivity measurements seemed to be effective for mapping of mobile and stagnant zones. Upon leachate recirculation a local decrease in average resistivity was observed;
- Ground penetrating radar (GPR) is another rapid developing technique for mapping subsoil in a non-destructive way. Results of ground penetrating radar were comparable to the electromagnetic conductivity measurements although penetration depth was less and costs were higher;
- CVES measurements (Continuous Vertical Electrical Soundings), also referred to as geo-electrical sounding or geo-electrical measurements, are measurements along a profile at the surface of a landfill. A geo-electrical survey, ultimately gives information on conductivity of the waste both at the surface and below. However, after having experience at four landfill sites it is believed that geo-electrical surveys might be able to detect areas of very low (< 7 Ωm), medium (10-30 Ωm) and high (> 20 à 50 Ωm) resistivity, which might be interpreted as respectively saturated zones with high salt contents (stagnant bulk); zones with low salt contents (mobile zones) and unsaturated zones. At Vlagheide, CVES-measurements were made before and after leachate infiltration, resulting in a decrease in resistivity around the place of infiltration. This may be contributed to an increase of contaminated water in this part of the landfill;

- The seismic tests (ConsoliTest™) uses seismic waves, created by hitting the surface with a sledge hammer. Seismic sensors on the soil record the vibrations and this gives information on mechanical properties (more or less the stiffness) of the first 10-30 meters below the surface. At the test sites, ConsoliTest™ results roughly confirmed EM and CVES measurements but spatial resolution is lower.

Tracer tests

In a tracer-test a certain amount of water is fed to the waste, with a tracer component added. This tracer is a component that normally doesn't appear in landfill leachate. Li or Br (sometimes combined LiBr), or certain organic components as fluorescein can be applied as tracers. The speed at which the tracer moves through the waste is measured, by taking water samples from sampling wells or from the leachate drainage system. Tracer tests can give quantitative information about residence-time distribution of water in the waste and on both the importance of preferential channels and possible mass-transfer into stagnant bulk.

Tracer tests are generally envisaged as a top-down test, where tracer is added at the top and extracted at the bottom. However, such a tracer-tests requires a very homogeneous infiltration and leachate collection systems, in order to give effective results and such systems are not available at most landfills.

For this reason an alternative is being developed in cooperation with University of Southampton, in which first tracer is added through the bottom-drainage system and subsequently extracted again with the same system, followed by model-interpretation of the results. A trial for this tracer test is in preparation on the Landgraaf landfill. Results are not yet available. Furthermore, volumes of water and chemicals involved are quite high (~1,000 m^3), therefore the test takes 2 to 3 months to run.

5. Sustainable landfill pilots

5.1 Monolith (A&G)

The monolith landfill concept is used for disposal of hazardous waste materials after stabilisation with a cement-like binder material. Stabilisation/solidification is a technology aimed at changing the physical/chemical properties of (hazardous) waste materials in order to reduce the release of contaminants in the environment. The technology aims at changing the release process from a percolation dominated to a diffusion or surface-dissolution dominated regime. For this form of treatment of hazardous waste for disposal in non-hazardous waste sites, the regulatory framework is still in development. In setting criteria for landfill classes in Annex II of the EU Landfill Directive (1999) it proved to be impossible to derive such values for stabilised monolithic waste due to lack of information on release and release controlling factors in stabilised waste monofills. Both at national and at EU level it was decided that additional information is needed to be able to develop proper criteria for this type of landfill. For the time being, regulatory controls were referred to the Member States. At this very moment a TAC working group is preparing recommendations for the EU Landfill Directive on a harmonised definition of monolithic waste, mechanical stability criteria and waste acceptance criteria.

The general scenario description for stabilized waste is given in figure 5.1. The scenario for stabilized waste is relatively complex in comparison with the scenario for granular materials. Many different chemical processes contribute to the potential emissions from the monolith. For a detailed description of the chemical/physical processes in a monolith landfill, the reader is referred to literature [3, 16, 33, 57-58, 65, 87, 89, 91-92, 94-95, 97].

The pilot experiment on the landfill site at “de Maasvlakte” in the Netherlands was terminated in 2007. Afterwards, the pilot experiment was carefully demolished to assess the physical structure of the monolith throughout the pilot. Samples were taken at different spots within the

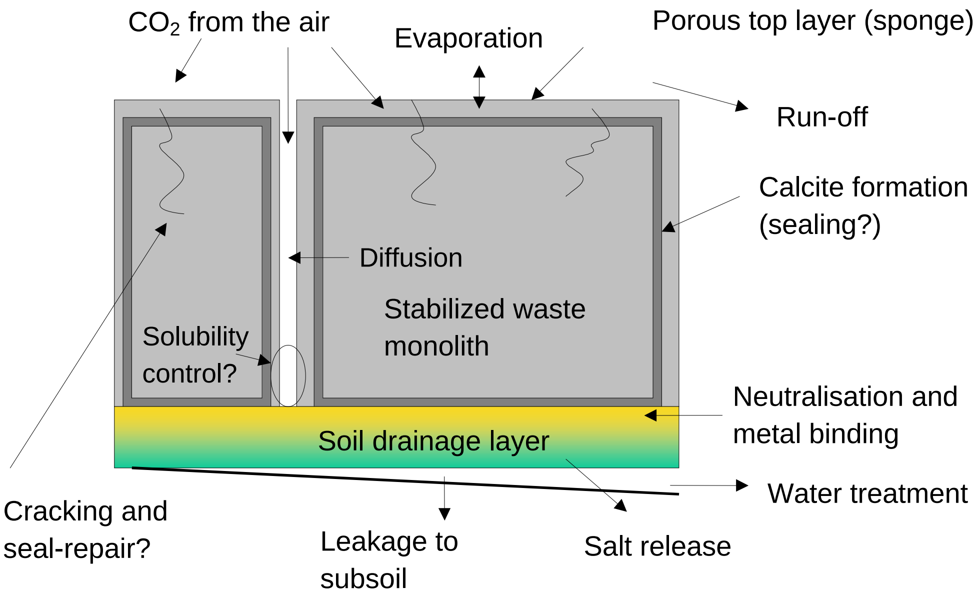

Figure 5.1 Scenario description for the monolith concept

monolith and analyzed for chemical/physical properties. Leaching experiments were also performed on the samples. One of the pilot cells was covered with an HDPE liner to prevent infiltration and intrusion of CO_2. The environmentally exposed stabilized waste cells were heavily weathered (change in color, cracking and swelling) up till 20-30 cm depth and lost part of the physical stability.

The unexposed cell (MSWI fly ash recipe wrapped in HDPE foil) was visually unchanged in comparison with the starting situation (no cracks or swelling). In addition, this material had been solidified and hence difficult to crush. The results from core samples clearly show the visual difference between the covered and uncovered cell and point at significant wash out of salts. These results are also consistent with the observation that the outer waste layer was substantially weathered and lost physical stability. Carbonation seems a secondary effect in the stability loss relative to salt release.

The Monolith pilot experiment has revealed several elements that might be of concern when no liner system is applied. The relevant elements are Br, Cl, Mo, Se and SO_4^{2-} [92]. As far as the risks of the monolith landfill at “de Maasvlakte” is concerned, it is questionable whether salts like chloride and sulphate pose a risk in this specific area close to the sea. With regard to establishment of European acceptance criteria for stabilized waste, it is important to note that differences exist in the environmental behavior between elements that are limited in their leaching by solubility control with mineral phases and elements that are not limited in their solubility (salts). The first group can be described by a source term accounting for the solubility limitation and sensitivity for pH changes in the material. The mobile salts exhibit diffusion controlled leaching and the source term needs to take diffusion processes into account.

5.2 Equifill (Nauerna)

The pilot project Equifill aims at creating a biogeochemical equilibrium between a landfill and the environment within a period of 30 years, in order to reduce the long-term risk and the aftercare needs. Over the past few years, the chemical processes leading to release of contaminants from the Equifill pilot experiment were studied [62, 68-73, 86-88, 91, 94]. The laboratory and pilot experiments on predominantly inorganic waste have revealed that the components chloride, sulphate and possibly antimony exceed the criteria specified in the Annex II of the landfill directive [1]. A major challenge is to develop means to predict the long-term leachate quality based on geochemical reaction transport modeling, taking into account the geochemistry, hydrological conditions and degradation of organic matter in landfills. The scenario description for the Equifill concept is given in figure 5.2. In this scenario, rain water infiltrates with a specific rate and reaches the mobile pores in the waste (preferential flow). Leachate leaves the landfill and is either transported to water treatment or infiltrates in the local subsoil. A point of compliance is defined at some distance from the landfill. At this point, the groundwater quality should meet certain requirements (ground- or drinking water quality).

A better understanding of interactions between contaminants and reactive surfaces in waste materials (e.g. natural organic matter, clay and iron/aluminium oxides) is of crucial importance for environmental risk assessment based on the described scenario (figure 5.2). Over recent years this understanding has significantly improved, resulting in “multi-surface” interaction models for ion adsorption onto iron/aluminium oxides and natural organic matter (humic- and fulvic acid). These models were implemented within the modeling framework ORCHESTRA [40]. The “multi-surfaces” modeling approach has been successfully applied to describe the speciation of metals in soils (e.g. [12-13]. Moreover, this modeling approach was recently used

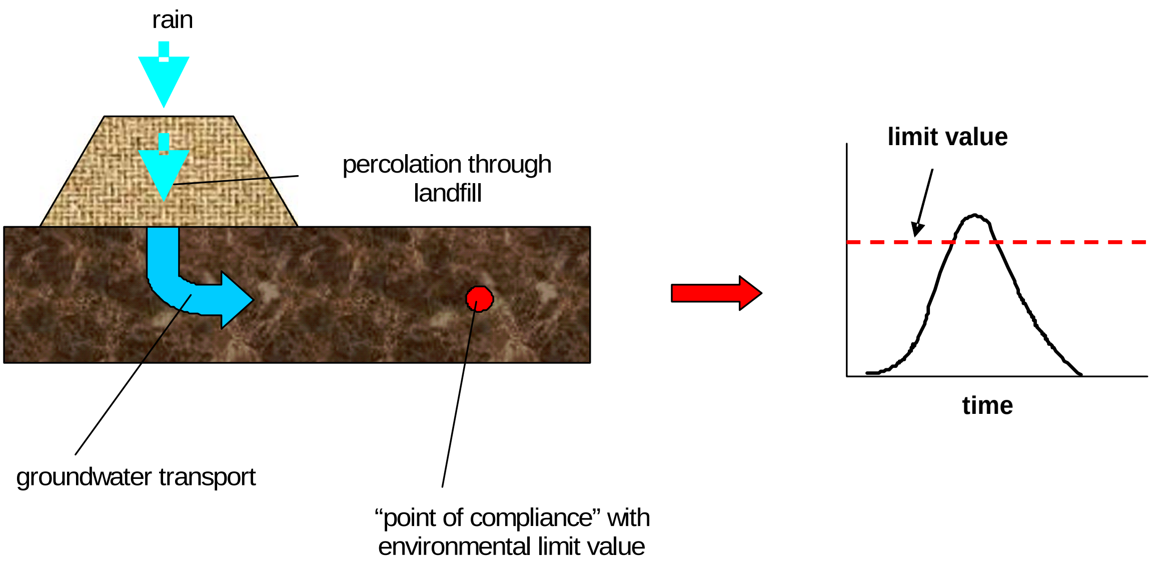

Figure 5.2 Scenario description for the Equifill concept

for derivation of new legislative emission values from construction products in the revision of the Dutch building materials decree [99-100].

This type of long-term prediction of emissions may be useful for decisions about the level of aftercare measures to be defined at the time of landfill closure. In a recent study, a new basis for determination of landfill after-care criteria based on site specific environmental risk assessment was presented [88]. The detailed modeling assessment is based on a source term, path and impact approach (as is shown in figure 5.2) and takes the chemical processes in the underlying soil system into account. This approach enables identification of possible critical contaminants based on a location specific environmental risk assessment. The model predictions showed that chloride, sulphate and possibly antimony will exceed drinking water quality criteria at 3m from the landfill. As such, these results are consistent with earlier conclusions about the risks of the Equifill pilot experiment [91]. The results implied that local environmental conditions contribute to the acceptability of landfill emissions and are important factors in choosing a landfill location. It was concluded that a risk-based approach for the evaluation of landfills could facilitate site-specific assessments of emissions from landfills to determine the degree of aftercare and/or decisions on the aftercare period.

5.3 Vlagheide

5.3.1 Introduction

Vlagheide in the south of the Netherlands measures 40 hectares and contains about 6 million m^3 of waste, stored over the last 40 years until December 2003. The older parts A and B of the landfill (22 ha) do not have a bottom liner, but are almost completely (for about 80%) sealed with an impermeable top-liner since 2002. The new part C of the landfill (18 ha) has an impermeable bottom liner, but is not yet capped with an impermeable top liner. Vlagheide is a representative example for hundreds of landfills in the Netherlands and Europe.

In 2005 project was started with the ultimate aim to implement and demonstrate strategies for sustainable and cost-saving aftercare, not being based on impermeable lining, but on control

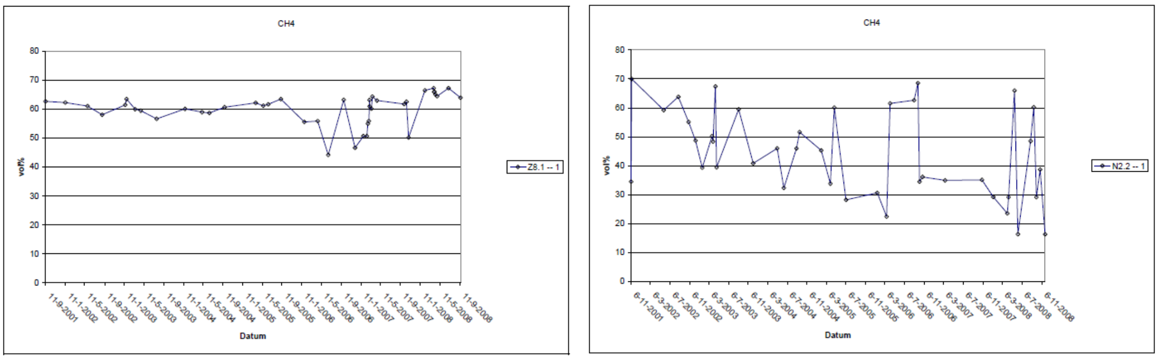

Figure 5.3 Methane trend '01-'08 without (left) and with (right) top liner

of natural biochemical processes in both the landfilled waste and the downstream polluted groundwater. The project is divided into three pilots:

- Pilot 1, situated at the old parts A and B of the landfill. The objective is to measure the differences between the part with a top liner (where infiltration of rain water stopped) and without the top liner (with infiltration of rain water);
- Pilot 2, situated in the groundwater underneath and downstream the old parts A and B. The objective is to measure the natural attenuation capacity in the contaminated groundwater plume, which is presently abstracted within the framework of a geohydrological containment measure. This pilot is less of importance of for this specific report on processes in the landfill itself and is not further reported here;
- Pilot 3, situated in the new part C of the landfill, where two compartments 4 and 3 were selected as a pilot and a control cell for leachate recirculation. The two compartments have similar waste-composition, similar composition of leachate and similar age of waste. The measuring period is 3 years.

The results are reported by van Vossen [77].

5.3.2 Results pilot 1 2005-2008 (effect top-liner)

Conclusions

The conclusions of pilot 1 are that there are clear and strong indications that the installation of an impermeable top liner reduces the occurrence of natural degradation in the waste body. This attributed to the fact that rainwater is prevented. An important indicator is biogas production, which dropped dramatically for the wells in the part that was covered (see Table 5.1).

In addition the methane concentration in the biogas produced, showed increased fluctuations together with a significant decrease over the period 2001-2008, as illustrated in figure 5.3.

Table 5.1 *Overview methane trends in gas extraction wells*

Position gas wells	number	*Distribution methane trends*							
			Strongly decreasing		Slightly decreasing		Stable		*Increasing*
		number	%	number	%	number	%	number	*%*
Without top liner	29	7	24%	12	41%	6	21%	4	*14%*
With top liner	20	17	85%	3	15%	0	0%	0	*0%*
At the edge of top liner	*6*	*3*	*50%*	*0*	*0%*	*2*	*33%*	*1*	*17%*

Drawing conclusions from biogas production values should be done with extreme care and any results are highly qualitative as the biogas production system consists of a distributed system of wells in which gas production is optimized. This means that air from outside the landfill is being drawn in at very different rates throughout the time of operation, causing unknown amounts of dilution.

5.3.3 Start-up of pilot 3 (infiltration-pilot)

Construction of the infiltration fields

Pilot 3 was started in the beginning of 2008 by installing 10 infiltration drains with a length of 100 meter each, separated at a distance of 10 m, in compartment 4 over a surface of 1 ha. The drains are situated in trenches back-filled with gravel. After a trial and error period, it appears to be possible to infiltrate a total amount of 1 to 1.5 m^3/hour which is roughly equal to an infiltration of 1,000 to 1,500 mm/year for the complete surface. This is four to five times more than the natural amount of rainwater that infiltrates in the waste body (estimated to be 300 mm/year).

Pilot 3 will last until the beginning of 2011, which means a total infiltration period of 3 years. First analysis of results took place about 4 months after start of the infiltration. At that time no clear increase of the gas production in the waste was observed, nor a change in the ratio BOD/COD in the leachate which is considered another a key performance indicator for the stabilization extent of the waste body.

The results from this pilot clearly show that measuring the impact of an infiltration experiment is not a straightforward operation. Special measurement wells and drains need to be installed to detect changes in biogas production. Additional processes need to be taken into account such as the impact the large increase in water flux has on the flow of gasses through the landfill.

5.4 TNO-CoFi: Development of a Risk Assessment Toolbox

As described in the earlier chapters of this report, most of the landfills pollution potential to soil and groundwater is governed by the (1) degree of biodegradation of the organic waste and (2) moisture distribution. For this reason a risk assessment toolbox is being developed[1], with the aim to come up with tools, which might be used to identify and characterize heterogeneity in the landfill body with the aim to identify the location of stagnant saturated or relative dry zones in the waste and quantify their importance. This project has finished in December 2009 and a final report is being written.

Since no single monitoring method available can give information on all these aspects of a landfill, risk assessment will most likely be based on a number of tools applied together, each providing one or more pieces of the puzzle. All pieces combined give a more detailed understanding of the processes occurring in a landfill and as such this understanding will improve future risk-assessment.

1 Project is partially funded by Stichting Duurzaam Storten. Other financers are Bodemzorg, Provincie Noord-Holland, Royal Haskoning and TNO.

A number of tools were applied to identify stagnant zones and to identify volumes of waste affected by preferential channels and mass-transfer from stagnant zones into preferential channels. The following tools were selected for testing, based on previous experience:

- Mapping of apparent electromagnetic conductivity in and surrounding the landfills (EM-31);
- Ground penetrating radar (GPR) survey;
- Resistivity profiling using geo-electrical sounding (Wenner electrode array);
- Induced seismic measurements of surface waves (ConsoliTest™);
- Cone-penetration tests (CPT) & borings and waste analyses (respiration or fermentation tests, leach-tests);
- Whole landfill tracer tests;
- High resolution sampling of monitoring wells and drains; with field measurements and laboratory chemical analyses;
- Molecular (DNA-)analyses of organic micro contaminants degrading organisms;
- Detection of stable carbon (^{13}C) isotopes.

Selected tools are evaluated in a series of trials at 6 different landfills in the Netherlands. During the project, research was continuously focused on most promising techniques. table 5.2 gives an overview of the different techniques applied in the project and the landfills where these techniques were applied.

5.4.1 Most important conclusions

Results from the project indicate that the following tools should be considered to be most promising for routine application as part of a risk assessment toolbox for landfills:

Electromagnetic conductivity measurements (EM-31) seem to be a quick and cheap additional technique to geo-electrical sounding and might be used as a first step in planning geo-electrical sounding campaigns. E.g., the method seemed to be effective for mapping of mobile and stagnant zones, as is illustrated with results of Vlagheide, where leachate was recirculated (figure 5.4). The local decrease in average resistivity, seen in both the EM and the resistivity data could be caused by leachate infiltration;

Table 5.2 *Research overview*

	Landgraaf	Bergen	Vlagheide	Kragge	N&M Zeeland	*Boom & Kemp*
Electromagnetic conductivity	X	X	X	X	X	*X*
Ground penetrating radar	X	X				
Geo-electrical sounding	X	X	X	X	X	*X*
Consilitest™	X	X				
Cone-penetration tests		X			X	*X*
Tracer Tests	X		X			
High resolution sampling	X	X		X	X	*X*
DNA-analyses		X				*X*
^{13}C isotope analyses		*X*				*X*

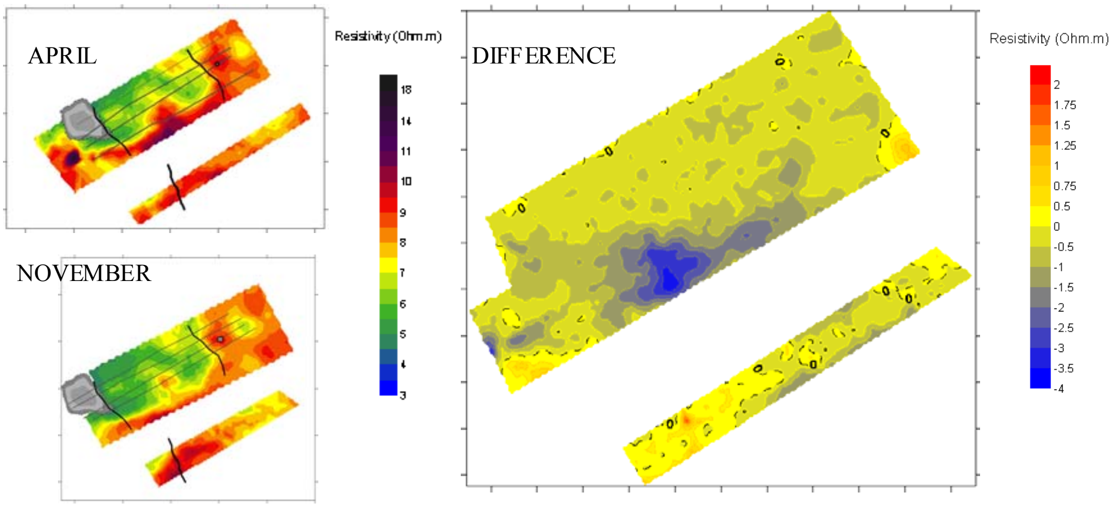

Figure 5.4 EM-31 image at Vlagheide. Left = before recirculation of leachate, April. Middle = after 5 months of recirculation of leachate, November. Right = difference between April and November. Grey area represents a pile of waste on which no EM-31 measurements were taken. Fat black lines indicate landfill compartments; parallel lines indicate location of geo-electrical measurements.

Geo-electrical sounding is normally used to assess resistivity distribution with depth in the subsoil. Resistivity is determined by water quality, waste composition and water content and therefore interpretation is difficult and often qualitative. The experience obtained at four landfill sites indicates that geo-electrical surveys give useful insights. At Landgraaf (figure 5.5) it seems than areas can be distinguished between very low (< 7 Ωm), medium (10-30Ωm) and high (> 20 à 50 Ωm) resistivity, which might be interpreted as respectively saturated zones with high salt contents (stagnant bulk); zones with low salt contents (mobile zones) and unsaturated zones. A few test-wells were available in the waste on or near the profiles and analyses of leachate-sample from those wells seem to confirm these conclusions.

Another application of geo-electrical sounding is to measure moisture content distribution. In other cases the objective was to evaluate the effectiveness of a water injection system [18-19, 52-53]. At the Vlagheide landfill geo-electrical sounding was performed before and after leachate infiltration, resulting in a decrease in resistivity around the place of infiltration (see figure 5.6). This may be contributed to an increase of contaminated water in this part of the landfill.

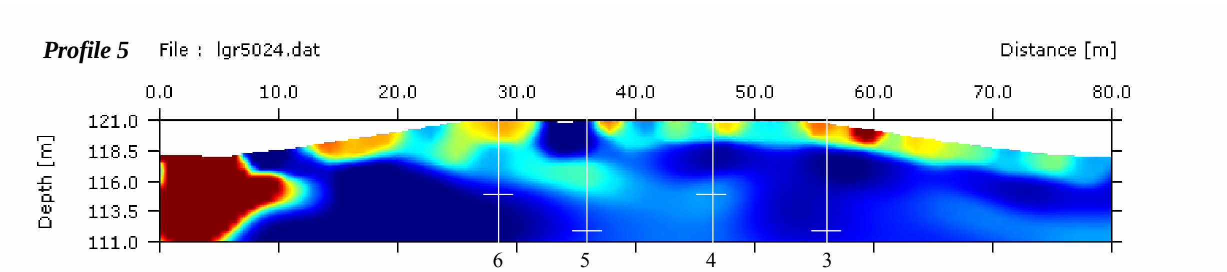

Figure 5.5 Sample profile of CVES-measurement at Landgraaf with test-wells on the edge of a low resistivity (dark blue) highly contaminated stagnant zone, and a higher resistivity (lighter blue) more mobile less contaminated zone, serving as a preferential channel (depth compared to NAP).

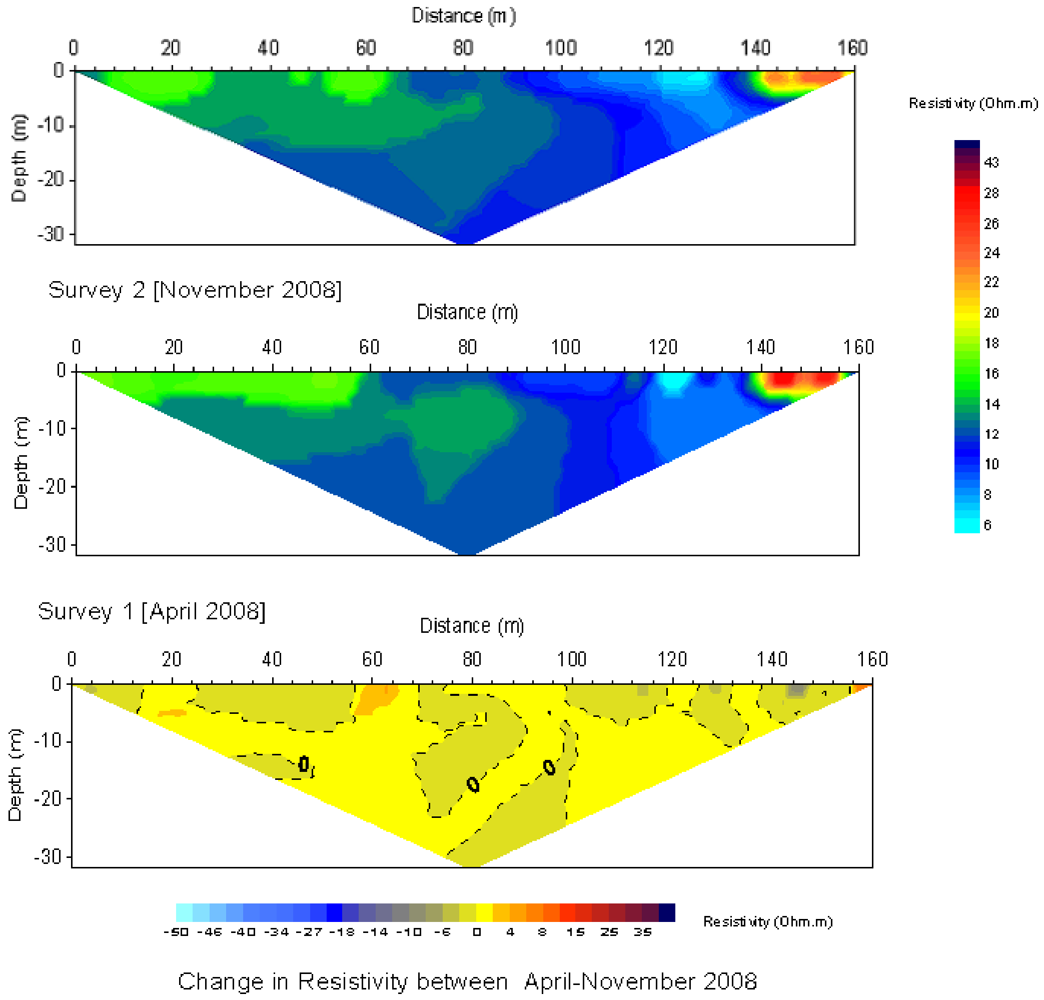

Figure 5.6 CVES geo-electrical-measurements at Vlagheide before (top) and after (middle) leachate infiltration, with a clear drop in resistivity in the infiltration zone. The lower image shows the difference between the two top images.

In Bergen, an attempt to link resistivity patterns to leachate quality failed, maybe due to quite homogeneous geochemistry within the landfill. However, resistivity measurements may be less adequate in saturated soil conditions. This aspect is subject of further research.

High resolution monitoring can be used to obtain a sharp picture of contaminant distribution throughout the waste disposal site. High resolution monitoring can be accomplished by using a fine-meshed well grid with multi level filters or an intricate drainage system on different levels. However a more extensive application might be possible by placing them, based on the results of geo-electrical sounding, e.g. in zones where sharp changes in resistivity are observed. In Bergen, DNA-measurements were performed in an attempt to find further prove for the hypothesized preferential biodegradation of aromatic compounds within the waste material. This additional evidence could not be found, probably due to interfering compounds. Stable ^{12}C and ^{13}C contaminant isotopes might provide important information on degradation processes, since ^{12}C containing compounds are preferentially degraded. In Bergen a decrease with depth was observed of the less degradable heavy benzene and naphthalene ^{13}C isotope fractions, indicating less degradation underneath the waste.

5.5 Proposed demonstration projects at De Kragge and Wieringermeer

5.5.1 Introduction

Within the framework the Dutch Sustainable Landfill Foundation the landfills “Kragge” and “Wieringermeer” were selected as locations for two demonstration projects, which will start after 2010. Numerous projects have shown that it is possible to apply technology to existing landfills in such a way, that long-term emissions are significantly reduced. The projects aim to provide an answer to the following questions:

- To what extent can we stimulate the natural bio-geo-chemical processes within existing landfills?
- Stimulation of natural bio-geo-chemical processes leads to a significant stabilization of the landfill body, but does it also lead to a significant reduction in potential emissions so that application of the technology will be economically feasible?
- Does this significant emission reduction also lead to acceptable emission levels given the current and future regulations?

The first step was to evaluate the technical and economical feasibility for projects on those two landfills. Results of this feasibility study are presented in a number of reports [78-82].

5.5.2 Kragge

General

Landfill “Kragge” is located near the city of Bergen op Zoom in the south-west of the Netherlands. Kragge is operated by Attero (formerly Essent Milieu) and was constructed in 1990. The 22 hectare landfill is divided into 5 waste cells. Compartments 1 and 2 have a surface sealing, compartment 3, 4 and 5 are covered with soil and MSWI ashes. A pilot for enhanced biodegradation is scheduled at compartments 3 to 5. Total area of the compartments 3 to 5 is about 11 ha. Total volume of waste in the cells 3 to 5 is 945.000 ton, distributed as follows over the different cells:

- cell 3: 300.000 ton (filled in the period 1997-2007);
- cell 4; 330.000 ton (2000-2007);
- Cell 5: 315.000 ton (2000-2008).

Height of the waste body ranges from 10 to 20 m. A gas drainage system with gas collectors and settlement beacons are present. The gas extraction system is operational since 1991. All compartments have a bottom liner, including a leachate drainage system.

Approach

The first phase of the project consists of a thorough pre-investigation program to characterize a.o. the actual degree of stabilization prior to the demonstration of stimulated biodegradation and as such emission reduction. Biodegradation will be stimulated by leachate infiltration, which will be operated for a number of years, until additional biogas production levels have become so low that stimulation of biodegradation is no longer feasible from a technical or economical point of view. Then, air will be injected to maximize biological stabilization by further oxidation. The duration of the leachate infiltration cannot be predicted at the moment. It might be somewhere between 4 and 10 years, depending on the current degree of biodegradation and the success of stimulating the anaerobic degradation by infiltration. The results of the pre-investigation program should provide the remaining gas potential of the

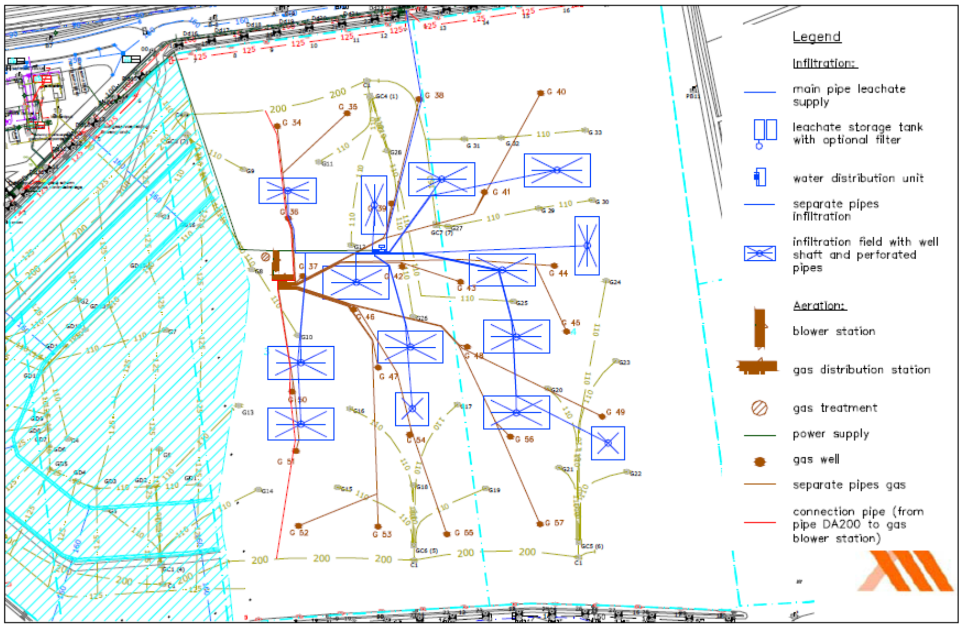

Figure 5.7 Proposed location of infiltration fields on top of Kragge

deposited waste and give a first indication of the duration of leachate infiltration. The aerobic in situ stabilization is expected to last at least 3.5 - 4 years, based on experience of aeration of landfill in Germany. After this period of time, a decision has to be made if continuation of aeration is required to reach acceptable emission levels. Careful monitoring combined with modeling should allow improved assessment of the effectiveness of the emission reduction efforts.

Design of the infiltration system

Leachate infiltration will be performed using infiltration fields below the top-liner system. Based on current experiences, this approach seems to be the best choice and is preferred above vertical infiltration wells or horizontal infiltration tubes, because it is demonstrated technology, relative cost-effective and durable.

Infiltration fields in the waste are generally shallow and wide pits (typically 1 m deep and 10-30 m wide), back-filled with gravel to which the leachate via a central pipe and some redistribution tubes. Infiltration fields are generally dug in just below the top-layer of the landfill, but might also partially be located on top of the waste. Infiltration fields with an automatic water distribution system were installed at the landfill Leppe and the landfill Mechernich in Germany.

Figure 5.7 shows the proposed location of infiltration fields on top of designated compartments. A total of 14 infiltration fields will be realized in the upper plateau area. They are positioned in a way that at one hand the influence on the waste package is maximized and at the other hand the risk of flooding the gas wells is reduced and instabilities of the slopes are avoided. The infiltration fields are supplied by a central water distribution station placed at the plateau of waste cell 4.

For the subsequent aeration-phase 24 new gas-wells will be drilled and combined with the existing gas-wells. Half of the wells will be used for air-injection, while the other half will be used for extraction. By setting the flow of extraction somewhat higher than the air-injection,

Table 5.3 *Primary and secondary key performance indicators (primary KPIs are listed in bold)*

General	Leachate	*Gas*
Temperature	**Redox (Eh)**	***Measured/calculated gas production***
Settlements	**Ammonia (NH4)**	***CH_4/CO_2 ratio***
Waste composition	**Conductivity (EC)**	***Gas extraction rate***
Moisture content	**Acidity (pH)**	*Oxygen (O_2)*
Moisture transport	**Biochemical oxygen demand (BOD)**	*Inhibitors*
Water balance	**Chemical oxygen demand (COD)**	
Time capsule	**COD-BOD**	
	BOD/COD ratio	
	Total organic carbon (TOC)	
	Dissolved organic carbon (DOC)	
	Benchmarking (a.o. LeachXS)	
	Chloride (Cl-)	
	Total Volatile Fatty Acids (VFA)	
	Alkalinity	
	Nutrients (including NA-parameters)	
	Sulphate (SO_4) and Sulphide (SO_2)	
	Nitrate (NO_3) and Nitrite (NO_2)	

undesired emissions are prevented. See for more details on such an air-injection system the description of the pilot at Wieringermeer in the next paragraph.

Monitoring

As concluded in chapter 0, many enhanced stabilization projects remain inconclusive because most attention focuses on technology for infiltration, while monitoring of effects is somewhat neglected. At Kragge therefore an intensive monitoring programme is defined, before, during and after enhanced stabilization. A summary is given in table 5.3. The preliminary conclusion based on leachate comparison is that landfill Kragge is further away from a stable situation than landfill Wieringermeer [81-82].

5.5.3 Wieringermeer

General

Landfill Wieringermeer is located near the town of Wieringermeer (20 km north of Hoorn) in the north-west of the Netherlands and is operated by Afvalzorg. The exploitation of the eastern part of the landfill has started 1985 and ended in 2000. The height of the eastern part of the landfill is 12 meters at the upper plateau. A project aim to stimulate emission reduction has been designed for compartment 6 on this eastern part. This compartment has a bottom liner and a leachate drainage system and settlement beacons. Gas is collected using 3 gas-wells. The cell is covered with a layer of soil varying from 1 to 1.5 m. Total area of the compartments 6 is about 2.6 ha. Volume of compartment 6 is 281,000 ton, of which 90% has been landfilled in the period 1992 -1994 and 10% in 1998.

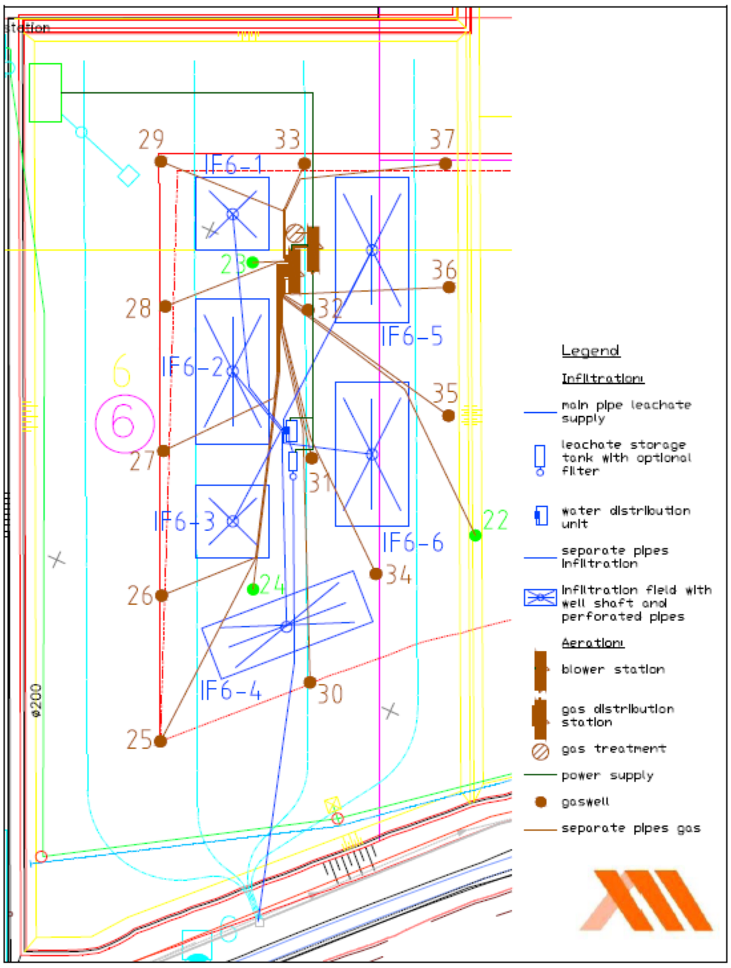

Figure 5.8 Location of gas injection wells and optional infiltration fields for Wieringermeer compartment 6.

Approach

The waste in Wieringermeer is older than waste at Kragge and results indicate that anaerobic biological stabilization has progressed to a much further extent than at the Kragge Landfill. Based on the results, additional stabilization of anaerobic degradation through infiltration is not considered to be cost-effective and further emission reduction should be achieved by commencing with aeration immediately. A pre-investigation program has been planned in order to determine the amount of biodegradable material left and the advantages and disadvantages of applying leachate infiltration as a first stage prior to aeration.

Design of the aeration system

For the aeration of compartment 6 13 new gas-wells will be drilled, with a depth of ca. 10 m and a diameter of 400 mm (see Figure 5.8). The 3 existing gas-wells (light-green) will be integrated in this system, yielding a total of 16 wells for aeration. In addition, HDPE-pipes will be dug into the top-liner system between the new gas-wells and the gas distribution station, for water supply to prevent the system from drying out.
Half of the gas-wells will be used for air-injection, while the other half will be used for extraction. From time to time, direction of flow will be altered, turning injection wells into extraction wells and vice versa. Total airflow is expected to be about 600 m^3/hr and by setting the flow of extraction somewhat higher than the air-injection, undesired emissions are prevented. After 3-4 years of aeration biodegradation is expected to be complete.

5.5.4 Monitoring

The aim is to carry out both the Kragge and the Wieringermeer demonstration projects at the same time. Both are focused towards the same goals:

- Assess the impact of technology to stimulate the bio-geochemical stabilization of waste with respect to reducing the long-term emission potential;
- Estimate the attainable emission-levels with respect to environmental regulations.

The demonstration projects must have an intensive monitoring program in order to be able to achieve these goals. We already know that the chosen technology for leachate infiltration and recirculation and landfill aeration will stimulate the bio-geo-chemical stabilization of waste, this has been shown in many projects throughout the world. The goal of nearly all these other projects, however, has never really been to reduce the long-term emission. In stead the goals were mostly focused on increasing the production of biogas, increasing biodegradation so that the available volume for waste storage can be more efficiently used etc. The long-term emission via leachate to groundwater is a very complicated issue which up till know has not been addressed.

The major reason for not focusing on long-term emissions via leachate in a quantitative manner has been the difficulty in coping with the heterogeneity of the landfill. As was mentioned in chapter 3, leachate flow through the landfill is a complex process where preferential flow plays a significant role in controlling emissions to the surrounding groundwater. We realize that no matter what technology we apply, we will never be able to completely reach a homogeneous situation in a landfill. In stead, we hypothesize that, in the stabilized landfill most easily degradable organic matter will have been degraded and the conditions will be anaerobic throughout the bulk of the landfill body. Stabilization, however, will result in geo-chemical gradients between the immobile anaerobic bulk and the much smaller fraction of preferential flow paths in the landfill which could be aerobic. Any degradable matter being released from the immobile bulk (by diffusion) will be degraded along the geo-chemical gradient before having the chance to migrate to the surroundings.

Main challenges for the monitoring are coping with the intrinsic landfill heterogeneity given the scale of the processes. Bio-geo-chemical processes occur at the pore-scale (in the order of cubic micrometers) whereas the demonstration projects are carried out at the scale of a landfill cell (in the order of thousands of cubic meters). In addition, sampling from the landfill and subsequent analysis in the laboratory will also be affected by these scale effects.
In order to cope with these issues so-called Key Performance Indicators (KPIs) were defined in the feasibility study. Three categories of KPIs were defined: general, leachate and gas. In addition a subdivision was made in primary KPIs providing direct information on the process of degradation of organic waste and secondary KPIs providing information on the required process conditions. Based on these KPIs, a proposal for a monitoring program and a preliminary investigation was made.

5.5.5 Plans for the pre-investigation program at Kragge and Wieringermeer landfills

For both landfills, all available data on waste composition, storage history, leachate and gas production have been collected and summarized in reports [81-82]. However some essential data on the current status are still missing. In order to acquire this information before the demonstration projects start, a pre-investigation program needs to be carried out. This investigation consists of the following steps:

- Sampling of the waste (approximately 24 samples from 12 drillings) in order to obtain additional chemical analyses and to carry out a series of tests on the samples. Analyses that are needed are amongst others water content, carbon content, characterization of organic fractions and degradability, and chemical composition. Tests will be carried out to quantify emission potential under aerobic and aerobic conditions and to quantify the biological activity present by respiration tests under aerobic and anaerobic conditions;
- Performing a series of aeration tests at existing gas wells. These tests are aimed to test the feasibility of injecting air into the landfill, to test the distribution of the air in the landfill and to see what effects air injection has on the landfill gas-emissions. These tests will be carried out with a mobile aeration system and will last a few weeks. Additional monitoring wells consisting of small pipes need to be installed in the vicinity of the wells where air is injected;
- Additional chemical analyses on leachate samples in order to acquire the required information for a complete analysis of the chemical speciation using the LeachXS expert system. This requires a full characterization of the macro-chemistry and the organic matter fractions in the leachate. It is essential to obtain an insight in the natural heterogeneity in leachate sampled from the drainage systems as well as leachate sampled from specific leachate wells.

5.5.6 Plans for the monitoring during the demonstration projects at Kragge and Wieringermeer

The monitoring program during the demonstration project is related to the KPIs given in Table 5.4. The frequency of monitoring depends strongly on the available budget. This monitoring program can be considered state-of-the-art in landfill applications. At the moment it is not yet clear if and how the proposed monitoring will lead to clear results on the effectiveness of the proposed measures to reduce the long term emission potential at both landfills. Although we have developed a pretty clear picture on what processes control the long-term emission potential in full-scale landfills, there remain major research questions concerning the impact landfill heterogeneity has on the long-term emission potential.

5.5.7 Fundamental STW research project proposal

Because of the problems related to the monitoring and assessment of the long-term emission potential of landfill bodies, a research proposal has been submitted to the Dutch Science Foundation to acquire additional funding for a more fundamental scientific research project. The ambition is to run this project in parallel with the demonstration projects.

This research project aims to develop an original and efficient measurement, monitoring and modeling framework for 1) quantification of the long-term emission potential of landfill bodies (with and without stabilization) and 2) optimization of the applied landfill stabilization technology for reducing the emission potential. Three sub-projects will be carried out:

- Integration of high resolution geophysical measurements with 3D process modeling to obtain 3D-time lapse images of processes in the landfill body;
- Quantification of bio-geochemical heterogeneous activity in full-scale landfills;
- Integrated modeling and up-scaling of landfill processes and heterogeneity.

The underlying assumption for this research project is that in order to obtain a quantitative understanding of the long-term emission potential data obtained from a landfill at a multitude of different scales should be integrated in a numerical model. Using a probabilistic approach which is tuned by the measured data will allow for a quantitative assessment of uncertainties which then can be used to assess the range of future emission-potential that is still present in the landfill body.

Table 5.4 *Monitoring program for the Kragge and Wieringermeer demonstration projects*

Item	Monitoring parameter	*Analysis frequencies*
Leachate	Flow/Volume	*Continuous in the leachate shafts (pump pits), on a daily to weekly basis*
	Composition1)	*6 times in the first year, then 4/year*
Infiltration medium	Composition1)	*6 times in the first year, then 4/year*
Infiltration system	Flow/Volume/Intervals for each infiltration field	*Continuous as part of system control, on a daily basis*
	Leachate level in system	*E.g. via vertical measurement wells in infiltration fields*
Landfill gas (before and during infiltration)	Flow/Volume	*Continuous via gas collection wells, at each gas well once a week to once a month*
	Composition2)	*Continuous via gas collection wells, at each gas well once a week to once a month*
	Gas temperature	*Continuous via gas extraction unit or gas recovery unit, at each gas well once a week to once a month*
Aeration / air supply (during aerobic in situ stabilization)	Flow, volume, pressure and temperature	*Continuous as part of system control, at each gas well once a week*
Extracted exhausts (during aerobic in situ stabilization)	Flow, volume, pressure and temperature	*Continuous via gas blower station as part of system control, at each gas well once a week*
	Composition2)	*Continuous via gas blower station as part of system control, at each gas well once a week*
	Leachate Level3)	*Continuous to monthly, e.g. via additional monitoring wells, optional in gas wells*
Waste body	Settlement/mechanical stability	*4/year, 50 m grid (settlement beacons, integration of existing beacons)*
	Water content	*Before start-up4)*
	Water storage capacity	*Before start-up4)*
Solid waste sampling in the waste body	Biodegradability, TOC	*Before start-up4)*
Meteorological data	*Temperature, atmospheric pressure, precipitation, atmospheric humidity, wind speed, etc.*	*Continuous on a daily basis*

1) pH, Conductivity, COD, TOC, BOD, TKN, NH_4, NO_3, NO_2, Cl, metallic compounds, phenols, phosphate, sulphides, AOX and relevant NA-parameters (leachate and infiltration medium might be identical).

2) CH_4, CO_2, O_2, H_2S.

3) Leachate level: especially examination of the free slopes when infiltration takes place within the area close to the slope.

4) Solid waste sampling before and after (and if necessary during) the controlled infiltration and aeration

6. Conclusions

6.1 Sustainable management of landfills and risk

As motivated in chapter 2, landfilling will remain an essential option of integrated waste management in order to treat considerable amounts of certain waste fractions and residuals of other waste treatment technology. The challenge we now face is how to integrate landfills in the total sustainable development framework of society.

It is becoming more and more apparent that integration of landfills in the sustainability framework requires a much broader assessment than one focussing only on environmental risk. Sustainability Potential Analysis, SPA, [32] is an approach based on the Function-Structure-Context framework. Here the functional requirements of a landfill are brought in to balance with structural and contextual issues. The functional requirements include elements like the capacity of a landfill to safely store the waste for a very long period of time. The structural elements include a description of the landfill system, the processes occurring within the landfill, the technology implemented at the landfill, the capacity of the landfill to keep on fulfilling its function after context changes, and the legislative and operational management procedures. Finally contextual elements include more or less everything beyond the physical and organizational boundaries of the landfill system and which may have an impact on the landfill. An important aspect of the SPA framework approach is the focus on obtaining a quantifiable assessment which allows a comparison between different scenarios.

The SPA approach can be a usefull instrument for assessing a landfill system as a whole. The SPA approach shows the necessity for integration of the currently available knowledge on landfill processes in order to predict the long-term emissions in a probabilistic sense.

6.2 Landfill processes

The mechanistic understanding of how landfill emissions develop and pose an environmental risk is gradually improving. To be more exact, we are developing important insights in the processes that are responsible for development of leachate and landfill gas. This knowledge has led to optimization of landfill gas production in order to maximize the energy production. Another important result is that we are more and more able to explain the lack of significant emissions resulting in pollution of soil and groundwater at many landfills. Because long-term potential emission via leachate is such a challenge this has been the main focus for this report. The emission of contaminants with leachate is ultimately the result of landfill hydrology, landfill bio-geochemistry and heterogeneity.

6.2.1 Landfill hydrology

It is obvious that flow of water is an important factor controlling landfill emission via leachate and this insight has lead to the construction of geotechnical barriers at landfills in order to prevent water flow as much as possible. However, recent scientific developments have provided much evidence that water movement is also an essential requirement for landfill stabilization which leads to reduction of the long-term emission potential.

Currently, the idea is that biological stabilization only occurs in zones with sufficient moving water. Monitoring of landfill hydrology clearly shows that water primarily flows through preferential flow paths which occur in an unknown but small fraction of the waste volume. Outside these zones with mobile water, water in most of the bulk waste will be stagnant and evidence has been found of even completely dry conditions in the landfill. In these zones without significant water movement, biological stabilization will occur at much slower rates that in zones with significant water movement. Exchange between the stagnant and mobile zones occurs through diffusion. This is a very slow process so contamination present in a stagnant zone will hardly contribute to pollution potential in the short term.

Understanding and controlling the flow of water through the body of a landfill is the key to the stabilization of the waste and therefore the reduction of the long-term emission potential. Enhancing biodegradation by infiltration and leachate recirculation has been adequately proven in laboratory- and pilot scale experiments. However, results from leachate infiltration and recirculation projects carried out at full-scale have been much less conclusive. This is attributed to the lack of high quality monitoring programs and as a result, high quality monitoring data [6].

Full-scale monitoring of landfill hydrology and leachate flow is a major scientific challenge, although significant progress has been made in the recent years. In the pilot projects carried out by DFSL in the first research phase (2000-2006), landfill hydrology has not been a specific focus of the monitoring effort. A review of the international literature also shows that not much research has been performed to investigate the landfill hydrology in an integrated framework together with the landfill bio-geochemistry and the landfill heterogeneity. In order to understand how manipulating landfill hydrology affects the long-term emission potential is an important research question that needs to be addressed in order to make the long-term emission potential reduction feasible and acceptable.

The concept for reduction of the long-term emission potential consists of initial stimulation of biodegradation processes by infiltration of water and subsequent leachate recirculation. This will significantly enhance landfill gas production. When gas production has dropped to low levels, at which landfill gas treatment becomes non-economical, leachate recirculation is stopped and the landfill will be drained and aerated as much as possible. Air can only move through the landfill pores that are not filled with water. Currently, not much is known about the heterogeneous distribution of water filled porosity in the landfill.

Quantitative modeling approaches (based on mechanistic principles), required for predicting long-term emission potential development and actual long-term emissions has yet to be developed. Development of these approaches requires additional research in order to understand the stability of the hydrology of the landfill over the very long time spans where landfills should maintain their function (centuries). Results from this research will help the development of technology to stimulate stabilization of much larger volumes of landfill waste in existing landfills and eventually make the implementation of SPA approaches for landfills in the Netherlands.

6.2.2 Landfill bio-geochemistry

Biodegradation of organic waste is the most important overall process taking place in the waste itself and this process controls the local chemical speciation of virtually all available chemical species present in the waste. Anaerobic biodegradation of organic matter in the waste is responsible for the production of landfill gas. Methane and carbon dioxide emissions have always played an important role in monitoring the behaviour of landfills. However, in the concept of reduction of the long-term emission potential, gas emission is a relatively short term process which should be manageable in a period of one or two decades. Landfill gas management has to be an important aspect related to the technology applied to reduce the long-term emission potential. Although gas production will continue because most of the landfill is in an anaerobic state, we expect that landfill gas emission is a much less important aspect in the long-term emission potential of a landfill because the gas production rate will become very small. Engineered bio-active aerobic layers can easily degrade the produced methane and therefore sustainably limit methane emissions in the long-term.

Biodegradation is also an important controlling factor for emission via the leachate. On the one hand, the complex range of microbiological reactions itself produces and degrades organic compounds which could add to the pollution potential themselves. On the other hand, bio-degradation influences DOC concentrations, pH and the redox status in the waste, essentially determining the release of heavy metals. The research carried out in the last decade has shown that control of DOC-levels in the leachate is one of the most important means for controlling the release of a host of substances of concern. Instead of focusing on analysis of trace organics, one should focus on bringing DOC levels down.

Although biodegradation at the pore-scale is not fully understood and much research effort is invested in this matter, the current state of knowledge seems to be sufficient to integrate biodegradation with the geochemistry of the waste in a quantitative manner. Since the first phase of the Sustainable Landfill project, significant steps have been made in understanding the key processes. The geochemical processes and controlling factors are pretty well understood by now. In spite of the huge complexity of waste and leachate, current computation and modeling technology can quantitatively predict emission levels of many macro elements and metals by simulating behaviour of many substances and many controlling factors simultaneously. An important challenge that we need to address is what consequences are of landfill heterogeneity on the long term emission potential. Field verification with long-term monitoring is crucial for validation of the model predictions. Not much research has been done on the fate of the organic micro-pollutants present in the landfill. However, biogeochemical reactivity of these micro-pollutants is known from soil contamination and integration in the current models should be relatively easy.

The concept of infiltration and leachate recirculation followed by an aeration phase will have a profound effect on the bio-geochemistry in the landfills. Air will flow through the preferential flow paths in the landfill. As a result the preferential flow paths will be oxidized whereas the bulk of the landfill will remain anaerobic. The presence of redox gradients along the diffusion path from the stagnant bulk of the landfill to the preferential flow paths is considered to be a benefit limiting the long-term emission potential. This combined with the presence of stabilized organic matter ensures that emissions will be low in a stabilized landfill. This assumption needs to be verified in the future.

For a monolithic waste landfill, the outcome of the demolition of the pilot experiment indicates that physical sustainability and stability is important for determination of the long-term emission of contaminants. It is concluded that a cover system for a stabilized waste landfill significantly increases the stability of the monolith due to decreased water infiltration and weathering processes of the material. The research challenge is to quantify permeability and stability of the landfill under field conditions over a long period of time. It is recommended to verify the development of cracking and strength by coring full scale stabilized waste disposal facilities at least 10 years after the start of the operation.

6.2.3 Landfill heterogeneity and scale

The experiences from the different projects carried out within the DFSL clearly indicate that landfill heterogeneity is a complicating factor for stabilization of waste. Heterogeneity of the waste materials in the landfill leads to preferential flow caused by a multitude of reasons including compacted layers with very low water permeability and the presence of waste contained in plastic bags. Another effect of heterogeneity can be seen on the scale of a toothpick, at the edge of the toothpick, hydrolysis occurs, leading to the growth of a bio-film in which the hydrolysis products are degraded to methane, carbon dioxide and water, the inside of the toothpick is left unaffected.

Another effect which plays an important role is the presence of geochemical gradients in the landfill which develop as the biogeochemical processes in the landfill unfold. Especially the expected redox gradient from the preferential flow towards the bulk waste in the landfill is expected to be important. To our knowledge no detailed research has been performed on this topic yet. Given the ambition of the DFSL to develop stabilization technology which has a significant impact on the redox state and distribution in the landfill, this research should have priority.

Currently no, truly integrated approach exists which adequately integrates these different scales for the assessment of landfill emission potential. Models have been developed that span all these scale ranges, some research groups have even attempted to integrate these scales in deterministic models, but the enormous amount of parameters that need to be quantified are a clear indication that this approach is not applicable for landfill management and assessment. Some alternative approaches have been attempted, but no satisfactory approach, able to provide quantitative predictions of leachate emission with reasonable accuracy has been developed yet. This is perhaps the most important challenge which needs to be solved for the integration of landfills in the context of sustainable development of society.

6.3 Landfill technology

6.3.1 Stabilization technology

There is a clear international consensus on how to enhance biological stabilization of landfill bodies. This can be done both under anaerobic conditions (through leachate recirculation) and aerobic conditions (through injection of air). Worldwide this has already been applied in 50 to 100 projects, and perhaps even more. Within the DFSL, two new pilots have been designed, based on a thorough review of these previous experiences. The main ambition in these two projects is to quantify the impact that these types of technology have on landfill stabilization and to what extent the long-term emission potential of the landfill is reduced. It is clear that success of these two new pilots will depend on an accurate assessment of the initial and final conditions, as well as on a detailed and accurate monitoring of the impact of the different technologies on the processes within the landfill. Proper monitoring of the effects of leachate infiltration and/or air injection proves to be a very difficult task and has more than once been underestimated.

Apart from the difficulties to prove enhanced biodegradation, it cannot be ruled out that actual enhancement of biodegradation is in fact unsatisfactory in many of the projects done worldwide. This might be attributed to an inadequate design of the system for infiltration or aeration or to insufficient amount of leachate infiltrated. It appears that preferential flow is a complicating factor, making it difficult to achieve flow in the immobile zones of the landfill. Considering the overview of knowledge on preferential flow, it now seems quite naïve to assume that addition of leachate to a landfill results in an increase in water content and thus increased biodegradation throughout the waste. Waste buried deep in the stagnant zones will be much more difficult to reach, and a more directed and sustained effort will be required to really promote biodegradation in those spots. Landfill aeration technology has shown to be able to degrade up to 90% of the degradable organic carbon present before aeration, so a combination of infiltration, leachate recirculation and aeration seems to be the optimal approach. Demonstration projects will need to be carried out in order to quantify the impact of this reduction on degradable organic carbon on the long-term emission potential of the landfill body.

It is clear that the stimulation of biodegradation will help to significantly reduce risks of landfills. Waste present deep into the stagnant zones in a stabilized landfill will have a very limited contribution to the emission with the leachate leaving the landfill because the presence of geochemical gradients and stabilised organic matter will absorb any mobile contaminants. The long-term mechanical stability of the landfill is an important issue which also needs to be taken into account in this assessment.

Clearly, it is of utmost importance to monitor hydrology, biodegradation and the heterogeneity of landfills in order to conclude on technical possibilities to reduce the pollution potential and to define even more effective systems for the future.

6.3.2 Monitoring

In the past years, a lot of effort has been put in the DFSL-projects to develop methods and approaches to understand the processes occurring within the landfill body. The ambition of these efforts was to reduce the environmental risk associated with emissions from the landfill. However, the DFSL projects were not focused on developing a risk assessment framework; this part still has to be done.

In order to obtain meaningful results from measurements and monitoring within the landfill we require a combination of different measurements and experiments at different scales. The experience in the DFSL projects has shown that interpretation of the results of a single landfill within an expert-system like LeachXS, based on state-of-the-art mechanistic models supplemented with a wide range of relevant data from a large number of other landfills, significantly improves the predictions that we can make with the data. In addition, our understanding of the status of a specific landfill is larger as well because monitoring data can be compared to data from other landfills.

Methods and tools are being developed so that we can understand the impact of landfill heterogeneity on our predictions and the development of the long-term emission potential. However, the current interpretation possibilities lead to a qualitative interpretation. The research community in the world is putting much effort in developing quantitative interpretation approaches. These quantitative approaches are based on integration of measurements at different scales with numerical models integrating the most important processes in a probabilistic sense. This knowledge needs to be integrated in the DFSL framework.

6.4 Recommendations for future research

The concepts developed for Sustainability Potential Analysis provide an excellent framework in which technology for long-term emission potential reduction can be integrated with risk analysis, life cycle analysis and other important contextual issues. The two demonstration projects that have been planned provide a unique chance to further develop the ideas presented in chapter 2.

Although much is known about the hydrological processes in the landfill, little mechanistic knowledge is available on the occurrence of preferential flow. Understanding the spatial and temporal distribution of preferential flow in a landfill is essential for assessing the long-term emission potential. This requires the further development of model concepts which need to be integrated with large scale monitoring and experimental concepts so that consequences of preferential flow at the scale of a complete landfill can be assessed in a probabilistic sense.

In addition to understanding preferential flow, it is essential to understand how diffusion from the bulk of the landfill waste towards the preferential flow paths leads to emission with leachate. High amounts of preferential flow combined with slow diffusion from the stagnant zone leads to low emissions with leachate. The consequences of preferential flow on the geochemical speciation of heavy metals, macro parameters and DOC need to be integrated in the modeling concepts that have been developed in the previous phases of the DFSL.

Sequential application of infiltration and recirculation followed by aeration technologies in landfills will increase the biogeochemical heterogeneity in the landfill because of the development of highly oxidized zones close to highly anaerobic zones. The current scientific agreement is that this is beneficial for reducing the long-term emission potential of a waste body. This has been confirmed by results from various projects. It now needs to shown that acceptable levels of remaining emission potential can be achieved and proven with sufficient reliability.

Monitoring and measurement tools need to be improved so that an integrated concept can be developed for assessing the development of the long-term emission potential at a landfill. This concept will inherently be a concept that copes with uncertainty by adopting a probabilistic approach in which models and measurement data are integrated. Integration of this approach with the LeachXS framework will give the added benefit of benchmarking a specific landfill against the behaviour of other landfills present in the database of LeachXS.

A major challenge is to obtain sufficient data from the demonstration projects at the Kragge and Wieringermeer landfills to unequivocally prove that the applied technology has led to a significant reduction in emission potential. Currently, an industry state-of-the-art monitoring program is planned, however, it is important to realize that the question being studied has up to now never been addressed in this fashion, and additional monitoring combined with modeling is required.

Application of geophysical techniques such as geo-electrical sounding and EM technology shows promising results. In order to move from the current qualitative interpretation towards a more quantitative assessment, the mechanistic concepts describing the processes occurring in the waste body need to be integrated in 3D models which then are then used to interpret the geophysical measurements.

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