Leachate management in the aftercare period of municipal waste landfills

WANG Yu





DOCTORAL DISSERTATIONS

Leachate management in the aftercare period of municipal waste landfills

WANG Yu

A doctoral dissertation completed for the degree of Doctor of Science in Technology to be defended, with the permission of the Aalto University School of Engineering, at a public examination held at the lecture hall R1 of the school on 3 May 2013 at 12.

Aalto University School of Engineering Department of Civil and Environmental Engineering

Supervising professor

Prof. Juha Kaila

Thesis advisor Acting Prof. Markku Pelkonen

Preliminary examiners

Prof. Zhao Qingliang, Harbin Institute of Technology, China Ass.Prof. Johann Fellner, Vienna University of Technology, Austria

Opponent

Prof. Peter Kjeldsen, Technical University of Denmark, Denmark

Aalto University publication series **DOCTORAL DISSERTATIONS** 71/2013

© WANG Yu

ISBN 978-952-60-5140-6 (printed) ISBN 978-952-60-5141-3 (pdf) ISSN-L 1799-4934 ISSN 1799-4934 (printed) ISSN 1799-4942 (pdf) http://urn.fi/URN:ISBN:978-952-60-5141-3

Unigrafia Oy Helsinki 2013

Finland

Publication orders (printed book): yu.wang@aalto.fi



441 697 Printed matter



Author

VANG Yu		
lame of the doctoral dissertation		
eachate management in the aftercare period of municipal waste landfills		
Publisher School of Engineering		
Jnit Department of Civil and Environmental Engineering		
eries Aalto University publication series DOCTORAL DISSERTATIONS 71/2013		
ield of research Waste Management		
Ianuscript submitted 2 November 2012 Date of the defence 3 Mathematical structure	ay 2013	
Permission to publish granted (date) 29 January 2013 Language	English	
Monograph Article dissertation (summary + original articles)		

Abstract

The selection of a landfill leachate management strategy in order to shorten the aftercare period and reduce the leachate management cost is challenging. For decision making, it is important to understand 1) the main indicators of long-term leachate performance, 2) the target levels of these indicators must reach to indicate the end of the aftercare period and 3) the strategy to meet the target level of the indicators within the shortest time. The aim of this thesis is to establish a leachate emission prognosis tool for the determination of the length of the aftercare period and to use models to test the effects of different leachate management strategies on the length and overall leachate management costs of landfill aftercare. The first part of the research is a study of municipal landfill stabilization and emissions, made by systematically describing the long-term landfill leachate and gas (LFG) emission performance achieved by landfill simulators (landfill simulation reactors, LSRs). The results give a comprehensive picture of the waste biodegradation progress during the landfill aftercare period. The second part of the research is an evaluation of the feasibility of a biological on-site process to pretreat the leachate (mainly total nitrogen [TN] removal) for leachate recirculation, direct discharge and indirect discharge purposes from both technical and economic points of view. It is integrated with a case study of a cost estimation based on a real landfill condition as an important part of the study, conducted to define the applicability of the crucial leachate management alternatives.

Based on the results of the LSR and biological leachate nitrogen removal studies, the possibility and feasibility of optimizing landfill leachate management and treatment were clarified by developing models for the estimation of long-term emissions from landfills of different sizes and evaluating the best options for leachate and nitrogen management during the aftercare period. The models developed can be used to express the importance of different target parameters and estimate the length of the aftercare period for a landfill that is effective at optimizing a cost-effective aftercare strategy. The modelling findings show that in conventional scenarios, without leachate recirculation rate, the aftercare period can be shortened substantially, to 25 years and 75 years, in medium-sized and big landfills respectively; though this is technically more challenging to do for big landfills. These scenarios also showed that the lowest total and average (per m³) leachate management costs can be achieved at about 60% of the costs of conventional scenarios during the aftercare period.

Keywords Municipal waste management, Landfill emissions, Leachate control and treatment strategy

ISBN (printed) 978-952-6	30-5140-6	ISBN (pdf) 978-9	52-60-5141-3	
ISSN-L 1799-4934	ISSN ((printed) 1799-4934	ISSN (pdf)	1799-4942
Location of publisher Es	spoo	Location of printing	Espoo	Year 2013
Pages 176		urn http://urn.fi/UR	N:ISBN:978-952	-60-5141-3

Preface

This dissertation is based on research carried out at the School of Engineering, Aalto University.

I am grateful to Professor Juha Kaila for his valuable advice, great encouragement and important guidance. He has made his support available in a number of ways; not only for research but also for personal career development related issues. Without his advice and unique support, this dissertation would never have been possible.

I would like to express my gratitude to acting Professor Markku Pelkonen for initiating me into scientific research and for having been such a helpful instructor during the past years. I appreciate his advice to commence the study and research into the area of waste and wastewater management and also value his inspiring ideas and advice throughout the research.

It is an honour for me to work in the Laboratory of Water and Wastewater Engineering and to know the great colleagues who have given me great support there. I would like to thank Professor Riku Vahala, Aino Peltola and Ari Järvinen for their invaluable help in both my life and the experiments.

I am indebted to my colleagues, Mari Pantsar-Kallio, Ari Makkonen and Jari Kuivanen, for their considerate encouragement and for good comments and discussions that have helped orientate me during the past years.

I owe my deepest gratitude to my parents for their support and belief in me. Finally, I wish to thank my wife Le for her inexhaustible love and patience with my studies and work.

Espoonlahti, October 2012

Wang Yu



Contents

Pr	eface	•••••		I
Co	ontents	•••••		
Li	st of Pu	ıblicatio	ons	VII
Au	thor's	Contri	bution	IX
Li	st of A	bbrevia	tions	XI
Li	st of Fi	gures		XV
Li	st of Ta	ables		XVII
1	Intro	ductior	Π	1
	1.1	Backg	ground	1
	1.2	Object	tives of the dissertation	2
2	Land	lfill and	l Leachate Management – A Literature Review	4
	2.1	Funda	mentals of landfill processes	4
		2.1.1	Introduction	4
		2.1.2	Landfill as a biological system	5
		2.1.3	Leachate and LFG	6
	2.2	Landfi	ill operational strategy	9
		2.2.1	Bioreactor landfills	9
		2.2.2	Studies on bioreactor landfills	11
	2.3	Leacha	ate in landfills	15
		2.3.1	The nitrogen cycle in landfill and leachate nitrogen	15
		2.3.2	Organics and inorganics of leachate	17
		2.3.3	Waste pretreatment impacts on leachate emission	

		2.3.4	Old and new landfills leachate situations and problems	
		2.3.5	Landfill temperature impacts on leachate emission	
		2.3.6	Landfill L/S ratio's impacts on leachate emission	
	2.4	Leacha	ate treatment	
		2.4.1	Leachate treatment alternatives	
		2.4.2	Nitrogen removal processes	
		2.4.3	Lab-scale and technical scale leachate treatment applications	
	2.5	Landfi	ll leachate management	
3	Mat	erials ar	ıd Methods	
	3.1	Landfi	11	
	3.2	Waste		
	3.3	Leacha	ate	
	3.4	Landfi	Il simulators (I, II)	
	3.5	Biolog	rical leachate treatment and cost estimation (III, IV)	
	3.6	Leacha	ate management scenario and strategy model (V)	
		3.6.1	Forecast modelling for target variables	
		3.6.2	Forecast modelling for the leachate emission integrator	
		3.6.3	Leachate management scenarios	
		3.6.4	Landfill leachate management strategy model	
	3.7	Landfi	Il leachate discharge limits	
4	Over	rview ar	nd discussion of results	52
	4.1	Leacha	ate emission and biodegradation of waste (I, II)	52
		4.1.1	Leachate emissions	
		4.1.2	Cumulative gas production and methane content	55
		4.1.3	Temperature impacts	56

	4.1.4	Discussion
4.2	Leacha	ate biological treatment: nitrogen removal (III, IV)
	4.2.1	Nitrification
	4.2.2	Denitrification
	4.2.3	Partial nitrification and nitrite denitrification test period
	4.2.4	Operational strategies
	4.2.5	Discussion
4.3	Cost es	stimation of leachate treatment (III, IV)
4.4	Landfi	ll leachate management: nitrogen control and treatment (V)
	4.4.1	Landfill emission potentials: NH4-N, COD, chloride and LFG emission 67
	4.4.2	Post-closure management: a leachate management scenario and strategic
	4.4.3	Discussion
Cone	clusions	

5



List of Publications

This dissertation consists of an overview and of the following publications that are referred to in the text by their Roman numerals.

- I Wang Y. and Pelkonen M., (2009) Impacts of temperature and liquid/solid ratio on anaerobic degradation of municipal solid waste: an emission investigation of landfill simulation reactors, Journal of Material Cycles and Waste Management, 11: 312-320
- II Wang Y., Pelkonen M. and Kaila J., (2012) Effects of temperature on the long-term behaviour of waste degradation, emissions and post-closure management based on landfill simulators, The Open Waste Management Journal, 5: 19-27
- III Wang Y., Pelkonen M. and Kaila J., (2011) Cost-saving biological nitrogen removal from strong ammonia landfill leachate, Waste Management and Research, 29 (8): 797-806
- IV Wang Y., Pelkonen M. and Kaila J., (2012) Strategies to enhance the biological nitrogen removal of high strength ammoniac and low C/N landfill leachate with SBR process, Environmental Technology, 3 (35): 579-588
- Wang Y., Pelkonen M. and Kaila J., (2012) Optimization of landfill leachate management in the aftercare period, Waste Management and Research, 30 (8): 789-799



Author's Contribution

The author has had an active role in all the phases of the research reported in this dissertation. He has participated in the planning and implementation of all experiments and developed most of the calculations and analysis of the results reported in publications I - V. He has also written the first drafts of the publications and revised them after comments by the supervisors.



List of Abbreviations

Ammonia (NH₃) Ammonium-nitrogen (NH4⁺-N) Anaerobic filter (AF) Biochemical methane potential (BMP) Biochemical oxygen demand (BOD) Carbon dioxide (CO₂) Carbon to nitrogen ratio (C/N ratio) Chemical oxygen demand (COD) Chloride (Cl⁻) Completely autotrophic nitrogen-removal over nitrite (CANON) Dissolved oxygen (DO) Deoxyribonucleic acid (DNA) European Union (EU) Flow injection analysis (FIA) Free ammonia (FA) Free hydroxylamine (FH) Granular activated carbon (GAC) Hydraulic retention time (HRT) Landfill gas (LFG) Leachate emission integrator (LE) Landfill simulator/Landfill simulation reactor (LSR) Leachate collection system (LCS) Liquid to solid ratio (L/S ratio) Methane (CH₄)

Mixed-liquor suspended solids (MLSS)

Municipal solid waste (MSW)

Nitrate (NO_3)

Nitric oxide (NO)

Nitrite (NO_2^{-})

Nitrogen dioxide (NO₂)

Nitrogen gas (N₂)

Nitrogen loading rate (NLR)

Nitrogen oxides (NOx)

Nitrous oxide (N₂O)

Not analysed (NA)

Not regulated (NR)

Organic loading rate (OLR)

Organic nitrogen (Norg)

Oxidation reduction potential (ORP)

Oxygen uptake rate (OUR)

Polymerase chain reaction (PCR)

Post-denitrification (post DN)

Putrescible fraction of municipal solid waste (PFMSW)

Ribonucleic acid (RNA)

Sequencing batch reactors (SBRs)

Simultaneous nitrification and denitrification (SND)

Sulphate reducing bacteria (SRB)

Total dissolved solids (TDS)

Total kjeldahl nitrogen (TKN)

Total nitrogen (TN)

Total organic carbon (TOC)

Total oxidized nitrogen (TON)

Total solids (TS)

Volatile fatty acids (VFAs)

Volatile solids (VS)

Wastewater treatment plant (WWTP)

Xenobiotic organic compounds (XOCs)

List of Figures

Figure 1 A schematic illustration of the thesis structure and the development steps of the
solution
Figure 2 Nitrogen flow from the environment to landfill
Figure 3 Atmospheric temperatures in the test landfill
Figure 4 A sample of landfill temperatures in the test landfill, 2003 – 2004
Figure 5 Precipitation in the test landfill area
Figure 6 Schematic diagram of a LSR
Figure 7 Schematic overview of a sequential batch reactor with activated sludge
Figure 8 Operational cycles of a sequential batch reactor with activated sludge
Figure 9 Leachate treatment options for cost estimation (*advanced treatment needed when
effluent COD does not meet the limit; ** \pm 25% unit cost changes used for cost estimation) 50
Figure 10 Variation of leachate NH ₄ -N, COD and Cl ⁻ as a function of temperature and the L/S
ratio
Figure 11 Modelled leachate concentrations and gas production
Figure 12 LE integrators at different temperatures
Figure 13 LE trends in different scenarios
Figure 14 Estimated total and average costs in different scenarios and leachate management
strategies (The vertical bars indicate the differences of the total costs, with \pm 25% unit cost
changes for on-site advanced treatment and the WWTP treatment fee considered in the cost
estimation.)



XVII

List of Tables

Table 1 Typical LFG composition	8
Table 2 Overview of the studies on bioreactor landfills	12
Table 3 Leachate concentration changes with landfill age	20
Table 4 Overview of the studies on landfill leachate treatment	29
Table 5 Mean characteristics (± standard deviations) of landfilled waste in the test landfill	36
Table 6 Mean characteristics of landfill leachate in the test landfill	37
Table 7 Thermal regime and waste charged in the LSRs	39
Table 8 Operational parameters of the LSRs	40
Table 9 Quality of raw influent landfill leachate (Not analysed [NA]) (Units: External car ml/day; others, mg/L)	bon, 43
Table 10 Operational parameters and influent (average COD, NH ₄ -N concentrations-stage	s) of
the sequential batch reactor with activated sludge (mean or mean \pm standard deviation)	44
Table 11 The general specifications of the six scenarios	49
Table 12 Major leachate discharge limit values in some countries, mg/L	51
Table 13 Temperature impact coefficients	58
Table 14 Coefficients of the leachate emission forecast models	68
Table 15 Weighting coefficients of leachate emission integrator	71

1 Introduction

1.1 Background

Historically, landfills were initiated largely as a result of a need to protect the environment and society from adverse impacts of alternative methods of refuse disposal, such as open-air burning, open-pit dumping and ocean dumping (Boni et al., 2006). Although landfills eliminated some negative impacts of the old alternatives, new problems arose, primarily due to gas and leachate formation.

In year 2008, around 93 million tons of municipal solid waste (MSW) was landfilled in European Union (EU) countries, accounting for about 43% of the total produced. This number had decreased by more than 30% compared with ten years earlier. However, the cumulative amount of waste landfilled in EU countries since the year 2000 exceeds 1000 million tons (European Commission, 2011). For example, in Finland the number of closed municipal landfills is over 2000, containing 20 - 35 million tons of landfilled waste. Meanwhile, in most developing and newly developed countries, landfilling will still be the dominant methodology for a long period. Landfills are usually operated for decades (e.g. 30 years is the period adopted in the operation of landfills in the US). In ideal scenarios, it is expected that landfills could biodegrade to stable conditions within 25 - 30 years after closure. However, in the real-scale landfills, especially in big landfills, the required liquid to solid ratio (L/S ratio) needed for biodegradation is difficult to achieve in this time, which means that the landfill aftercare period is quite long and costly (Christensen et al., 1992). Compared with a 50-year aftercare period landfill, the operational cost, i.e. the landfill gas and leachate emission control cost and the landfill cover and piping system maintenance cost, will be high if this period is prolonged to 100 years.

For decision making concerning landfills, it is important to understand the long term potential environmental emissions and to predict the length of the aftercare period. The three aspects to consider in regard to this are 1) the main indicators of long term leachate performance, 2) the target level for these indicators to reach to indicate the end of the aftercare period and 3) the strategy to meet the target level of the indicators with optimized total management costs.

In order to achieve the sustainability of landfill operations, there is a need to develop a prognosis tool that enables the evaluation of the leachate emission level and prediction of the length of the aftercare period with the designed landfill leachate management strategy. The strategy directly affects the capacity of designed landfill leachate management facilities. The evaluation of the landfill emission level needs to address different aspects significant to environmental and economic issues. However, little research has been conducted evaluating the crucial elements that affect optimization of aftercare strategies in view of the sustainability of closed landfills (Laner, 2011).

1.2 Objectives of the dissertation

The challenges of landfill leachate management in the aftercare period raise questions like:

- What is the length of the landfill aftercare period from the point of view of leachate emission limits, leachate characteristics and changing trends in the aftercare period?
- Can specific leachate treatment strategies be implemented to affect leachate management, e.g. leachate recirculation in the aftercare period from environmental and economic perspectives in different types of landfills?

Hence, the major aim of this thesis is to develop a leachate emission prognosis tool for the determination of the length of the aftercare period and to use models to test the effects of different leachate management strategies on the length and overall leachate management costs of landfill aftercare.

The analysis in this study concentrates on the environmental and economic aspects of closed landfills; the social dimensions of long-term landfill management are not addressed.

The research scope of this thesis can be categorized into the following (Figure 1):

- Leachate characteristics and emission potentials in the aftercare period: systematically
 describes the long-term landfill leachate and landfill gas (LFG) emission performance
 achieved by landfill simulators (landfill simulation reactors, LSRs), which gives a
 comprehensive picture of the waste biodegradation progress and its interrelated effect
 on the long-term landfill emission potential during the landfill aftercare period
 (publications I and II).
- Developing leachate treatment strategy to improve leachate management in the aftercare

period: The leachate management principles and strategies are developed from the technical and economic evaluation of leachate management methods, i.e. an on-site process to pretreat the leachate (mainly total nitrogen [TN]) for recirculation, affecting the length and costs of leachate aftercare (publications III and IV).

 Methodology verification and validation: based on the results of LSRs and biological leachate nitrogen removal studies, the leachate emission prognosis model for determination of the target L/S ratio values and estimation of the length of aftercare period was developed. With the developed model, a scenario-based methodology is used to assess the leachate management principles and strategies (publication V).



Figure 1 A schematic illustration of the thesis structure and the development steps of the solution

2 Landfill and Leachate Management – A Literature Review

2.1 Fundamentals of landfill processes

2.1.1 Introduction

Until the middle of the 20th century, waste was simply heaped on the ground, and the disposal site was called a dump (Nathanson, 2003). These uncontrolled, open dumps quickly posed serious environmental concerns including offensive odours, unsightly nuisances, pollution of groundwater and surface water, and potential fire risks. Open dumping of solid waste material is no longer an acceptable disposal method in the EU countries and in many other countries.

On the other hand, most municipal waste is now treated in a sanitary landfill, not simply deposited in a pile on the ground (Jokela et al., 2002a; Nathanson, 2003). The municipal sanitary landfill is distinguished from the open dump because the solid waste is placed and compacted in a suitably selected and prepared landfill site in a carefully prescribed manner with a daily layer of compacted soil (Nathanson, 2003).

In the recent past, it was believed that a suitable depth or thickness of naturally occurring soil between the bottom of the landfill site and the groundwater table or bedrock would suffice to prevent pollution. But, by the 1980s, it was discovered that soil does not necessarily absorb or attenuate all the contaminants seeping out from a sanitary landfill, no matter how thick the underlying soil layer. In order to prevent potential groundwater pollution, suitable bottom liners and leachate collection systems (LCSs) are required at new landfill sites; these two systems are major components of a well-engineered landfill site. Most new landfills are constructed with composite liners, which collect the leachate for centralized treatment prior to disposal.

With increasing knowledge of waste biodegradation in landfill sites, moisture content has been claimed to play an essential part in stabilization of biodegradable organic fractions (El-Fadel et al., 1997a; El-Fadel et al., 2002; Sormunen et al., 2008). In order to increase the biological degradation rate and reduce the aftercare period, the operation mode of landfills has undergone a dramatic change, evolving from a storage/containment concept to a process-based approach; in other words, to a bioreactor landfill (Yuen, 2001; Warith, 2002). The conventional landfill approach refers to the placement of waste into landfills with lined bases and caps, intended to

preserve the waste in a relatively inactive stage by minimizing the amount of water entering into the waste mass in order to reduce the production of LFG and leachate (Yuen, 2001; McBean et al., 2007). However, there is a long-term risk that the suppressed biodegradation may turn active in the future when the containment system becomes aged, and thus having the long-term potential of causing serious environmental damage (Yuen, 2001). In recent years several research studies have demonstrated that the application of landfill as a bioreactor could be viable (Komilis et al., 1999a, 1999b; Benson et al., 2007; Sormunen et al., 2008; Gioannis et al., 2009). A bioreactor landfill is an engineered landfill that enhances the biodegradation of waste by recirculating some amounts of leachate or adopting other methods including waste shredding, waste compaction, pH adjustment and aeration, as well as the addition of nutrients and increase in alkalinity (Reinhart et al., 2002).

Although landfills eliminated some of the negative impacts of the old alternatives (open-air burning, open-pit dumping and ocean dumping), new problems arose, primarily due to gas and leachate formation. There are three major alternatives for the treatment of landfill leachates; leachate channelling, biological treatment and chemical/physical treatment. In addition, several alternatives exist to reduce the emission of LFGs, including passive oxidation in the soil top covers, combustion in flares and LFG utilization for energy generation (as electricity or cogeneration). Markedly, the latter saves emissions to the environment, because emissions are avoided that would have occurred if the same amount of electricity/heat produced from LFG had been produced from fossil resources (Manfredi et al., 2009a).

2.1.2 Landfill as a biological system

Despite landfilled solid waste composition varying substantially, for a lot of reasons (socioeconomic conditions, waste collection strategies and waste disposal methods), the total organic content constitutes the highest percentage of solid waste (Boni et al., 2006). The composition of organic matter in landfills is site-dependent as well. For example, food waste constitutes over 60% of the household waste in China while it is around 25 - 30% in Finland. Additionally, municipal solid waste in Finland has a higher proportion of garden waste, whereas the waste in China contains little such waste. Most organic materials can be broken down into simpler compounds, leading to the formation of gas and leachate. These conversion processes are mainly performed by the bacterial activity of aerobic and anaerobic micro-organisms; recent studies have proven that there are a variety of bacterial species involved in the degradation processes of waste solids in landfills (Boni et al., 2006).

In an anaerobic landfill, a multi-step process including hydrolysis, acidogenesis, acetogenesis and methanogenesis may happen. Under anaerobic conditions, organic matters are decomposed sequentially into complex particulate matters and then into simpler polymers like proteins, carbohydrates and lipids. The polymers are further hydrolyzed and finally converted into either intermediate by-products, acetic acid or hydrogen. Methane is primarily generated by acetate decomposing and then carbon dioxide reduction with hydrogen (El-Fadel and Massoud, 2001). For example, under anaerobic conditions, the cellulosic material is converted to methane (Zehnder, 1978) by the hydrolytic and fermentative bacteria (from polymers into sugars), the obligate hydrogen reducing acetogenic bacteria (from sugars into acetate and hydrogen) and the methanogenic bacteria (finally converting it into methane). In the presence of sulphate, the sulphate reducing bacteria (SRB) may be active in the degradation process with the production of hydrogen sulphide and carbon dioxide (Barlaz et al., 1992).

Aerobic bioreactor conditions result in organic nitrogen, such as protein, is hydrolyzed and fermented to NH_4^+ . The nitrogen content of municipal waste is less than 1% on a wet-weight basis – mainly from yard wastes, food wastes, and biosolids. If dissolved oxygen and sufficient alkalinity are present, NH_4^+ is further oxidized by nitrification into NO_3^- , resulting in the destruction of alkalinity and a drop in the pH.

2.1.3 Leachate and LFG

Leachate is formed when the waste moisture content exceeds its field capacity. Moisture retention is attributed primarily to the holding forces of surface tension and capillary pressure (El-Fadel et al., 1997b). Percolation carries soluble compounds through the refuse mass, resulting in highly polluted leachate; soluble compounds are generally encountered in the refuse at emplacement or are formed in chemical and biological processes. The sources of percolating water are primarily precipitation, irrigation and runoff, which cause infiltration through the landfill cover; initial moisture and newly produced water due to refuse biodegradation may also contribute to leachate formation but in smaller amounts (El-Fadel et al., 1997a). Increased moisture content not only significantly contributes to leachate formation, but also enhances biodegradation processes in landfills, resulting in reduction of the time required for the return of the landfill to beneficial land use.

The quantity of leachate generated is site-specific and determined by water availability and weather conditions as well as the characteristics of the waste, the landfill surface and the underlying soil. In relatively warm climates, for example, the increase in leachate production after precipitation is generally quite rapid; in colder climates, it is common for leachate production to lag behind precipitation because a large part of it falls as snow. In addition, leachate production is generally greater whenever the waste is less compacted, since compacting reduces the filtration rate (Lema et al., 1988; Hossain et al., 2003).

The quality of landfill leachate is highly dependent upon the stage of fermentation in the landfill, waste composition, operational procedures and co-disposal of industrial wastes. The leachate contains four groups of pollutants: dissolved organic matter, inorganic macrocomponents, heavy metals and xenobiotic organic compounds (XOCs). Although it is difficult to generalize concerning the particular chemical concentration that a leachate will contain, the trend of a continually decreasing concentration is a generally observed phenomenon. In the acid phase, concentrations are generally higher due to enhanced formation of dissolved organic matter and the release of ammonium. In the methanogenic phase, the content of dissolved organic matter significantly decreases and the composition of the organic matter changes, indicated by a BOD/COD ratio below 0.1. The ammonium concentration seems not to follow the same decreasing trend and may constitute one of the major long-term pollutants in landfill leachate. The content of heavy metals in the leachates is generally very low as a result of attenuating processes (sorption and precipitation) that take place within the disposed waste.

Within a time horizon of five centuries, a larger fraction of the organic matter would probably leave the system through emissions of gas and leachate, but heavy metals would still be stored in the landfill (Manfredi and Christensen, 2009b). Boni et al. (2006) reported the total amounts of heavy metals released during the experimental activity represented only a very limited fraction (in some cases, lower than 10%) of the initial content measured in the bulk waste.

Leachate contains a broad variety of XOCs. The most frequently observed compounds are aromatic hydrocarbons and chlorinated aliphatic compounds. The XOCs are affected by volatilization and degradation as well as leaching and are, in most cases, not believed to be a long-term problem (El-Fadel et al., 1997a; Haarstad and Mæhlum, 1999; El-Fadel et al., 2002; Kjeldsen et al., 2002).

When waste is deposited in a landfill, biodegradable organic materials react quickly with the available oxygen to form carbon dioxide, water and other by-products (e.g. bacterial cells) as

part of aerobic decomposition. The anaerobic decomposition phase starts as oxygen depletes within the landfill. Although a landfill ecosystem undergoes an initial, short aerobic decomposition phase, the subsequent anaerobic phase is the dominant phase in the landfill's timespan (El-Fadel et al., 1997a).

Under a stabilized methanogenic condition, which is the stage of interest from a beneficial recovery perspective, methane and carbon dioxide are by far the two principal components of LFG and form more than 90% of the total gas generated. Nitrogen and oxygen are normally present in small quantities primarily as a result of air entrapment during waste deposition, atmospheric air diffusion through the landfill cover (especially in the near surface layers) or air intrusion from negative landfill pressure when LFG is extracted. Table 1 summarizes the composition of a typical LFG. Besides potential adverse health effects and environmental pollution, trace compounds, even at low levels, could cause toxicity on microbial populations and hence may inhibit gas formation and stabilization processes within a landfill (El-Fadel et al., 1997a).

Table 1 Typical LFG composition

Source:	El-Fadel	et al.,	1997a
---------	----------	---------	-------

Component	Concentration Range Percent Dry Volume Basis
Methane	40–70
Carbon Dioxide	30–60
Carbon Monoxide	0–3
Nitrogen	3–5
Oxygen	0–3
Hydrogen	0–5
Hydrogen Sulphide	0–2
Trace Compounds	0–1

Many factors affect the gas generation rate including waste characteristics, the moisture content, the temperature, the pH, the availability of nutrients and microbes, and the presence of inhibitors (Boltze and de Freitas, 1997). The large size of pieces of MSW is suspected to

decrease the ability of microbes to degrade the material. The effect of particle size reduction was initially explained by the increase in the surface area available for microbial attach. According to Ferguson (1993), the surface area increases only slightly with decreasing particle size for fibrous particles, such as paper. Thus, the effect of size reduction on the methane production in landfills may be that relatively large pieces of plastic, paper or other materials shield the materials beneath them from infiltrating water. The shielded waste may remain too dry for biodegradation. Shredding breaks down the impermeable barriers and more of the waste is exposed to water (Ferguson, 1993; El-Fadel et al., 1997a).

2.2 Landfill operational strategy

2.2.1 Bioreactor landfills

In recent years, the operation of landfills as bioreactors has been confirmed as an effective way to enhance stabilization, mainly because leachate recirculation increases the moisture content and stimulates the microbial activity by providing better contact between insoluble substrates, soluble nutrients and micro-organisms (Komilis et al., 1999a, 1999b; Benson et al., 2007; Giannis et al., 2008; Sormunen et al., 2008; Gioannis et al., 2009).

The enhanced biodegradation can significantly reduce the required time for waste stabilization and, by successive re-uses of the same bioreactor landfill cell, there are overall savings arising from not requiring new landfill sites every 15 - 20 years (Warith, 2002; Reinhart et al., 2002). Bioreactor landfills can be operated under aerobic anaerobic or combined conditions. In fact, there is no entirely aerobic condition in landfills due to the non-uniform distribution of air, water and waste; in aerobic bioreactor landfills there are some anaerobic pockets throughout the waste mass (Berge et al., 2005, 2006). Operational conditions affect the organic matter degradation pathways and rates, the quality of leachate, potential environmental pollution and the operational cost (Barlaz et al., 1992; Berge et al., 2005; Giannis et al., 2008). According to different operational conditions, bioreactor landfills can be divided into four types: anaerobic, aerobic, facultative and hybrid systems. Under anaerobic conditions, there is no degradation pathway for ammonium, resulting in accumulated ammonium in leachate with high potential environmental toxicity; however, operational costs required by air addition can be saved and methane can be captured and reused (Berge et al., 2005). Aerobic bioreactor landfills degrade organic compounds (mostly converting them into carbon dioxide) in shorter time periods compared to anaerobic landfills as a result of elevated temperatures; in situ nitrification is feasible in an aerated solid waste environment and the potential for simultaneous nitrification and denitrification is improved because of the presence of both aerobic and anoxic areas (Berge et al., 2005; Berge et al., 2006; Giannis et al., 2008). At the same time, some chemicals that do not degrade or transform under anaerobic conditions may do so under aerobic conditions (Reinhart et al., 2002). Hybrid bioreactor landfills are operated on a pattern of alternating aerobic and anaerobic conditions via sequencing of air injection throughout the life cycle of the landfill; this specific degradation condition can serve to treat organic material more completely and favour several nitrogen removal processes. Lastly, facultative bioreactor landfills are operated with the intent of actively degrading the waste mass and, at the same time, controlling the high ammonium nitrogen in leachate is converted to nitrite/nitrate in an external treatment system prior to recirculation, then the nitrogen content is reduced through denitrification when recirculating nitrate and/or nitrite abundant leachate back into the landfill (Berge et al., 2005).

Waste moisture content is a crucial factor in the design and operation of bioreactor landfills. Leachate introduction techniques include surface application and injection through vertical wells or horizontal trenches, whereby leachate is recirculated through the waste, as opposed to it being treated and released into the environment. Generally, a moisture content of about 40% is essential for rapid aerobic degradation; microbial activity will be slowed down by several orders of magnitude if the moisture content is lower than 40% (Giannis et al., 2008). Operating the landfill as a bioreactor provides opportunities for in situ nitrogen transformation and removal processes. When adding air to landfills, biological processes such as nitrification can be enhanced because air diffusion may now occur within the waste mass. Additionally, recirculation of the pretreated (nitrified) leachate will improve the denitrification processes occuring in anoxic areas (Berge et al., 2005). Due to the heterogeneous nature of waste (in regard to temperature, oxygen levels and moisture contents), within one landfill cell there may be many nitrogen transformation processes occurring simultaneously or sequentially. These processes may include biological nitrogen conversion mechanisms such as ammonification, nitrification, denitrification, anaerobic ammonium oxidation and nitrate reduction (Berge et al., 2005). Mertoglu et al. (2006) found nitrification activity in the aerobic bioreactor and this was proven by the identification of large amounts of Nitrosomonas-like ammonium oxidizers and Nitrospira-related oxidizers with 16S rDNA and amoA based molecular microbiology techniques. It should be noted that it is impossible to ensure fully aerobic conditions throughout the entire landfill, but what is important is that the conditions within the site are as close to ideal

as possible for the majority of the site. Nitrification and denitrification may occur simultaneously in an aerobic bioreactor. Some research has been conducted evaluating the potential processes of nitrogen transformation and removal in landfills and it is meaningful to have a deep understanding of the processes that can be achieved and used to optimize the operation of bioreactor landfills.

2.2.2 Studies on bioreactor landfills

There have been several reports on bioreactor landfills under aerobic, anaerobic or combined conditions at lab-scale and field scale, as well as various process-based stabilization enhancement techniques (Reinhart, 1996; Warith et al., 2005; Benson et al., 2007; Sormunen et al., 2008). An overview of the studies is summarized in Table 2 and the details are discussed hereunder.

Mehta et al. (2002) investigated waste decomposition in the presence and absence of leachate recirculation in a field-scale landfill. After about 3 years of operation, waste was excavated from three enhanced cells (E1, E2 and E3) and two control cells (C1 and C2). It was found that leachate recirculation resulted in an increase in the waste moisture content, but that the waste in the enhanced cell was not uniformly wet. Leachate recirculation resulted in both higher methane yields and increased settlement. The extent of decomposition of excavated waste samples was determined by the biochemical methane potential (BMP) and the ratio of cellulose plus hemicellulose to lignin. The average BMP in the enhanced and control cells were 24.0 and 30.9 mL CH_4/dry g and the corresponding ratios were 1.09 and 1.44 respectively.

Jiang et al. (2007) studied the performance of pilot-scale anaerobic bioreactor landfills with different leachate recirculation volumes. Leachate recirculation with a high rate of 5.3% (v/v) per week can be adopted as an effective approach to remove organic pollutants in leachate. However, this may result in large amounts of organic matter being washed out of the landfill by the leachate, reducing methane production. Therefore, the level of leachate recirculation should be adjusted, depending on the different waste biodegradation phases, to achieve the dual purpose of high efficiency energy recovery and organic pollutants removal.

Authors	Application	Comments
Mehta et al. (2002)	A field-scale landfill in the presence and absence of leachate recirculation	Leachate recirculation can improve waste moisture content, methane yields and waste settlement
Warith (2002)	An experimental-scale LSR and field-scale landfill	Bioreactor landfills allow a more active landfill management that recognizes the biological, chemical and physical processes involved in a landfill environment. The effect of solid waste particle size, leachate recirculation and nutrient balance on the rate of MSW biodegradation were studied and analysed.
Jokela et al. (2002a)	Lab-scale LSRs	The methane and leachate emission potential of various MSW fractions, as well as the effects of aerobic treatment on the pollutant potential, were discussed.
Sponza and Ağdağ (2004)	Lab-scale LSRs in the presence (recirculation rates: 13% and 30% of the reactor volume) and absence of leachate recirculation	A recirculation rate of 13% of the reactor volume resulted in better leachate emissions (e.g. chemical oxygen demand [COD], volatile fatty acids [VFAs]) and LFG etc. but was not impactful on ammonium.
Sanphoti et al. (2006)	A simulated landfill with leachate recirculation in tropical conditions.	Leachate recirculation and supplemental water addition resulted in better performance in terms of cumulative methane production and the stabilization period required.
Jiang et al. (2007)	A pilot-scale landfill with different leachate recirculation volumes	Leachate recirculation volumes should be adjusted with the different biodegradation phases.
McBean et al. (2007)	A demonstration-scale bioreactor landfill	Biogas generation rate and waste settlement were studied respectively.
Benson et al. (2007)	Full-scale bioreactor landfills	The effect of leachate recirculation on leachate generation rates was found to be little but rather affected leachate quality.
Giannis et al. (2008)	Long-term lab-scale aerobic bioreactor simulators	Over 90% COD and biochemical oxygen demand (BOD) reductions were achieved and a low BOD/COD ratio of 0.017 showed the simulator reached a stable state. Simultaneous nitrification and denitrification were observed from the effective nitrogen reduction after the adjustment phase.

Table 2 Overview of the studies on bioreactor landfills

Sponza and Ağdağ (2004) studied the effects of leachate recirculation and the recirculation rate on the anaerobic treatment of domestic solid waste in three simulated bioreactors, one without and the other two with leachate recirculation. The change in the recirculated leachate volume from 9 L (13% of the reactor volume) to 21 L (30% of the reactor volume) was selected and the

pH, COD, VFA concentrations, methane gas productions and methane percentages in the bioreactor with 9 L recirculated leachate were better than bioreactors with 21 L and without leachate recirculation. However, leachate recirculation was not effective in removing ammonium from the leachate.

A high temperature results in a large loss of moisture in landfill refuse via evaporation and leachate recirculation may be insufficient to maintain the moisture content; supplemental water addition into the landfill is then necessary to reach the required moisture value. Sanphoti et al. (2006) investigated the effects of leachate recirculation with supplemental water addition on waste decomposition in tropical landfills. Anaerobic digestion with leachate recirculation and supplemental water addition allowed for the highest performance in terms of cumulative methane production and the stabilization period required. It produced an accumulated methane production of 54.87 L/kg dry weight of MSW at an average rate of 0.58 L/kg dry weight/d and took 180 days to reach the stabilization phase. However, the reactor with only leachate recirculation produced 17.04 L/kg dry weight at a rate of 0.14 L/kg dry weight/d and reached the stabilization phase on day 290. In comparison, the control reactor provided 9.02 L/kg dry weight at a rate of 0.10 L/kg dry weight/d, and reached the stabilization phase on day 270.

Benson et al. (2007) analysed five bioreactor landfills to provide the performance of bioreactor landfills operating in North America by comparing with conventional landfills. Leachate generation rates and leachate depths in landfill LCSs appear no different in bioreactor and conventional landfills, as are the leachate and liner temperatures. With respect to leachate quality, bioreactor landfills generally produce stronger leachate during the first 2 - 3 years of recirculation. Thereafter, leachates from conventional and bioreactor landfills appear to become similar. While the BOD and COD decreased, the pH remained around neutral and ammonium concentrations remained elevated. After about three years of recirculation, the BOD/COD ratio began decreasing appreciably. One year later, the BOD/COD ratio reached approximately 0.1.

Biodegradation of organic material in a landfill is usually stimulated when the water content reaches up to 50 - 70% due to leachate recirculation (Lema et al., 1988; Jiang et al., 2007) and methane production at the initial moisture content is 0.0003 - 4.5% in a big landfill and 0.2 - 1.0% in a medium-sized landfill, while at 60% moisture content methane production was 2.3 - 34% and 0.9 - 32% of BMP in big and medium-sized landfills, respectively (Sormunen et al., 2008).

Warith (2002) reported the effect of solid waste size, leachate recirculation and nutrient balance on the rate of MSW biodegradation. It was found that the smaller the size of the MSW, the
faster the biodegradation rate of the waste, and addition of primary sludge can result in a rapid decrease in BOD and COD concentrations in the effluent samples because primary sludge is a good source of microbial inoculums. When recirculating leachate into the landfill, the average pH of the leachate in the early stages of recirculation was on the acidic range of the pH scale; however, the pH value was in the range of 7 - 8 after two years of leachate recirculation.

Giannis et al. (2008) reported long-term biodegradation of MSW in an aerobic simulated landfill bioreactor during 510 days of operation. After 70 days of treatment, there was a COD and BOD₅ reduction of 78% and 97% respectively, while by the end of the experiment it had reached 90% and 99.6% respectively, with a low BOD₅/ COD ratio of 0.017 suggesting the aerobic reactor had reached a stable state. Results suggested that nitrification and denitrification occurred simultaneously. The NO₃⁻ concentration increased periodically and took 240 days to reach the maximum 33.5 mg/L. Thereafter it started decreasing and, at the end of the experiment, it was 10 mg/L.

McBean et al. (2007) described the performance of a 6200 ton demonstration-scale anaerobic bioreactor, with respect to the biogas generation rate and waste settlement. The methane generation potential was determined to be 167 m³/ton of waste and the methane generation rate constant was estimated as 0.51 per year. Landfill settlement over a 28-month period, ranged from 4% to 19%, with an average 11% subsidence across the landfill. Subsidence occurred rapidly in the first eight months of operation and then decelerated.

Jokela et al. (2002a) investigated the methane and leachate emission potential of various MSW fractions produced in source-separation and mechanical and biological pretreatment processes, as well as the effects of aerobic treatment on the pollutant potential. The untreated putrescible fraction of municipal solid waste (PFMSW) had a high methane yield and NH₄–N leaching potential, whereas the composted putrescible fraction of municipal solid waste (CPFMSW) produced less, and the emission potential of ammonium and total nitrogen from grey waste (the residues after recyclables and biowaste collection) was estimated to be at a similar level to that of CPFMSW. Aeration in lysimeters reduced CH₄ potential by more than 68% for the PFMSW and CPFMSW samples, whereas for the lysimeter landfilled grey waste the reduction was 50%. The effective separation and biological treatment of PFMSW can result in reducing the environmental impact of waste disposal in landfills.

The data above illustrates the potential of bioreactor landfills to enhance settlement, methane production and solids decomposition on a field-scale. However, high recirculation rates in full-

scale landfills may cause some operational problems, such as ponding or clogging, especially in areas with high precipitation (Jiang et al., 2007). High recirculation volumes may deplete the buffering capacity and remove the activity of methanogens. Therefore, a leachate management strategy for recirculation, e.g. the volume and strength, should be carefully considered and studied in order to maintain the benefit and eliminate the disadvantage of leachate recirculation; an optimum leachate recirculation volume can contribute to a COD decrease and effective methane gas production. In most of the studies referred to, the leachate management strategies for recirculation purposes during the aftercare period have not been addressed before, indicating the need for additional studies.

2.3 Leachate in landfills

2.3.1 The nitrogen cycle in landfill and leachate nitrogen

About 0.02% of the global nitrogen that is biologically available is an essential element of life and is ranked as the most important element after carbon, hydrogen and oxygen (Jokela and Rintala, 2003). Only a relatively small number of organisms are able to utilize N₂ in the process of nitrogen fixation due to the high amounts of energy required to break the triple bond of N_2 (Brock and Madigan, 1991). Most living organisms, including humans, adopt more easily available forms of nitrogen, which are ammonium and nitrate. Nitrogen in the mass of living organisms is mostly bound up in amino acids in the form of proteins or in the form of organic nitrogen, which can be incorporated as nucleic acids in deoxyribonucleic acid (DNA) and ribonucleic acid (RNA) (Jokela and Rintala, 2003). Presently, 40% of nitrogen is fixed by natural sources and 60% by human-derived sources. Obviously this has drastically increased the amount of the more easily bio-available forms of nitrogen (e.g. ammonium and nitrate), resulting in increased production and utilization of nitrogenous fertilizer for food production (Galloway 1998). Consequently, a lot of the food produced ends up as solid waste and wastewater treatment produced sludge, which are frequently disposed of in landfills. Figure 2 represents the nitrogen flow from the environment to waste management (landfill) (Jokela and Rintala, 2003).

The efficiency of nitrogen in food products versus its input as fertilizer, or as human-induced biological nitrogen fixation, in the EU has been estimated to be between 20% and 30%; with 70

- 80% of the rest going into the soil or air during cultivation (Isermann and Isermann, 1998). MSW has been estimated to be about 4% protein, mostly from food products. Via protein conversion, the process of ammonification happens to generate ammonia, which is finally dissolved in the leachate (Berge et al., 2005). It is also possible that the ammonia generated within landfills may combine with organic matter (i.e. carboxyls, quinine hydroxyls). Ammonia is stable under anaerobic conditions, which typically accumulates in leachate (Berge et al., 2005).



Figure 2 Nitrogen flow from the environment to landfill

Source: Jokela and Rintala, 2003

In an aerobic landfill, when air is added, ammonia can be converted into nitrate and/or nitrite via biological activity. The nitrate and nitrite produced can be further reduced to nitrogen gas by denitrification occurring in the anaerobic conditions (Giannis et al., 2008). The related contents are discussed in the section on bioreactor landfills. In bioreactor landfills, because nitrous oxide is a potent greenhouse gas, its production is a concern. Nitrous oxide is produced by partial denitrification and can also be a by-product of nitrification with a low partial pressure of oxygen (Berge et al., 2006).

2.3.2 Organics and inorganics of leachate

Organic components in leachate can be divided into two groups: dissolved organic matter and XOCs (Kjeldsen et al., 2002). Although there is little information on the composition of the dissolved organic matter in landfill leachate, several bulk parameters (COD, BOD and total organic carbon [TOC]) are usually used to cover a variety of organic degradation products, ranging from small volatile acids to refractory fulvic- and humic-like compounds. Volatile acids are usually accumulated during the acidogenic phase whereas fulvic- and humic-like compounds are the main pollutants of methanogenic leachate. On the most general level, the BOD/COD ratio is adopted to indicate the biodegradability of dissolved organic matter in leachate.

The most frequently found XOCs are the monoaromatic hydrocarbons (benzene, toluene, ethylbenzene and xylene) and halogenated hydrocarbons, such as tetrachloroethylene and trichloroethylene. XOCs originate from household or industrial chemicals and are present in relatively low concentrations of less than 1 mg/L of individual compounds (Kjeldsen et al., 2002). These compounds can also be harmful in small concentrations through their acute toxicity, low degradation rate, high bioaccumulation and chronic effects, e.g. on the hormone composition in humans and reproducibility (Haarstad and Mæhlum, 1999). The old MSW landfills usually have a higher level of XOCs than the newer landfills.

Heavy metals are the main constituent of inorganic matter in leachate. Fe, Cr, Cu, Mn, Mo and Zn are part of necessary biological processes; other metals, such as As, Cd, Pb and Hg, are considered toxic, even in very small concentrations. The content of toxic metals like Cd, Cr, Zn, Hg and Pb can be 100 times higher in landfills than in natural soils (Haarstad and Mæhlum, 1999). However, most heavy metal concentrations in landfill leachate are at or below the US drinking water standards (Kjeldsen et al., 2002). Heavy metal balances for landfills have shown that less than 0.02% of heavy metals received at landfills are leached from the landfill after 30 years; both sorption and precipitation are believed to be significant mechanisms for the metal's immobilization and the subsequent low leachate concentrations (Kjeldsen et al., 2002). Leachate recirculation reduces metal concentrations in leachate due to sulphide and hydroxide precipitation and reaction with humic-like substances (Warith et al., 2005).

Other inorganic matter comprises of calcium, magnesium, sodium, potassium, ammonium, iron, manganese, chloride, sulphate and hydrogen carbonate. Chloride is a very conservative contaminant and it would pass through older layers of the landfill without any significant

attenuation. Calcium and sulphate concentrations' decrease with time was attributed to the depletion of these compounds and to the pH increase reducing their solubility in leachate and enhancing precipitation (El-Fadel et al., 2002). Leachate recirculation maintains neutral or above neutral conditions, as well as stimulating reducing conditions favourable for the reduction of sulphate to sulphide (Warith et al., 2005).

2.3.3 Waste pretreatment impacts on leachate emission

The EU strategy for waste management strives for the sustainable use of natural resources, emphasizing waste prevention, material recycling and energy recovery. Since the adoption of the EU waste policies, including the EU framework directive on waste and directives on hazardous waste, packaging and packaging waste and landfill of waste (European Commissions, 1975, 1991a, 1991b, 1991c, 1994, 1996, 1997, 1999), the waste management practices of the EU member states have been subject to major changes.

The Landfill Directive (European Commission, 1999), the Directive 2000/76/EC on waste incineration (European Commission, 2000) and the packaging and waste packaging directive (European Commission, 2004) are the basis of the current European policy on waste (Buttol et al., 2007). Waste minimization and pretreatment before landfilling are encouraged, which in turn affects the composition of landfilled waste. The quality of landfill leachate is highly dependent upon the stage of fermentation in the landfill, waste composition, operational procedures and co-disposal of industrial wastes. So, the adoption of new EU directives on waste management will have some impact on leachate quality. Solid waste pretreatment techniques are traditionally associated with mechanical, thermal and biological pretreatment. Baling is a mechanical alternative for waste pretreatment that reduces the moisture retention capability of the waste, hence baled waste starts producing leachate earlier than expected. Leachate from baled waste has lower pollutant concentrations and cumulative organic leaching. However, baling processes might retard biodegradation due to reduced moisture flow and hindered nutrient and biomass exchange (El-Fadel et al., 2002).

Aerobic pretreatment prior to landfill also has several impacts on leachate: 1) aerobic pretreatment removes readily decomposable matter. Thus, a balance between the acidogenic and methanogenic stage in landfill occurs and little to no inhibition of methanogens takes place; 2) aerobically pretreated MSW acts as a diluent for the organic acids produced, preventing consequent methanogenic inhibition due to the low pH; 3) aerobic pretreatment increases the

temperature of the waste, making conditions favourable for enhanced methanogenic activity; 4) partial decomposition of lignin during aerobic conditions makes cellulose and hemicellulose, the primary substrates during anaerobiosis, more readily available for methanogenic conversion (Komilis et al., 1999; Berge et al., 2005). Moreover, anaerobic mechanical-biological pretreatment is an alternative applied in central Europe, for instance, in Germany etc.

Moreover, recycling paper and inorganic components can reduce the total leachable COD by 25% and iron loading by 80% per unit of waste landfilled compared with unsorted refuse. Removal of recyclables in combination with aerobic pretreatment can result in a more than 90% reduction of both COD and iron leachable loading compared with leachate from unsorted MSW (Komilis et al., 1999).

2.3.4 Old and new landfills leachate situations and problems

The main difference in organic matter between old and new leachates is that old leachate has a low percentage of readily biodegradable material due to a considerable percentage of slow biodegradation components in final leachate effluent (Kjeldsen et al., 2002). Slow biodegradation components are complex organic compounds such as humic substances and XOCs. Leachate composition concentration variation with landfill age is represented in Table 3 (Farquhar, 1989; Laner, 2011). The data show magnitudes because climate and landfill size etc. have a great effect. It is observed that the ammonium-nitrogen concentration tends to be higher in the leachate from bioreactor landfills than from conventional landfills. In bioreactor landfills, the rate of ammonification is increased by moisture addition, even after the biodegradable organic fractions of the waste are removed (Berge et al., 2005). A landfill site may still produce leachate with a high concentration of NH₄-N over 50 years after filling operations have ceased. If not properly treated, the leachate that seeps from a landfill can enter the underlying groundwater environment posing a potentially serious hazard to public health because of its aquatic toxicity and oxygen demand in receiving waters. Therefore, the treatment of leachate to remove ammonium is an important aspect of long-term landfill management (Price et al., 2003). In addition, although the biodegradability of leachate organic compounds declines with time, complex organic compounds remain in solution. Thus, old leachate requires an adjusted and integrated alternation of biological, physical and/or chemical treatment processes to reach discharge limits. It is likely that ammonium-nitrogen and organics are the major compounds to determine when the landfill is biologically stable and when post-closure monitoring may end (Berge et al., 2007).

Table 3 Leachate concentration changes with landfill age

Sources: (Farquhar, 1989; Laner, 2011)

Parameter (mg/L)	Landfill leachate age (year)							
Turumeter (mg/L)	0-5	5 - 10	10 - 20	>20				
BOD	800 - 15 000	200 - 3 000	100 - 1 000	<100				
COD	3 000 - 30 000	1 000 - 15 000	1 000 - 5 000	<1 000				
TN	1 000 - 3 000	400 - 2000	75 - 500	<200				
Nitrogen (ammonium)	500 - 3 000	300 - 2000	50 - 500	<200				
TDS	10 000 - 25 000	5 000 - 10 000	2 000 - 5 000	<1 000				
pH	3-6	6-7	7 – 7.5	7.5				
Calcium	2 000 - 4 000	500 - 2 000	300 - 500	<300				
Sodium and potassium	2 000 - 4 000	500 - 1 500	100 - 500	<100				
Magnesium and iron	500 - 1 500	500 - 1 000	100 - 500	<100				
Zinc and aluminum	100 - 200	50 - 100	10 - 50	<10				
Chloride	1 000 - 5 000	500 - 4 000	50 - 1000	<200				
Sulphate	500 - 2 000	200 - 1 000	50 - 200	<50				
Total phosphorus	100 - 300	10 - 100	NA	<10				

2.3.5 Landfill temperature impacts on leachate emission

The quality of leachate varies significantly between acidogenesis and methanogenesis, so any factors affecting the waste biodegradation, such as methane production and the transition from acidogenesis to methanogenesis, will have great impacts on leachate quality. The microbial degradation rate increases along with temperature increase. In the suitable temperature range, an

increase of 10 °C will double the biodegradation rate. Rees (1980) observed that the optimum temperature for methane production from domestic waste in a conventional anaerobic digester is about 40 °C. Similarly, Hartz et al. (1982) investigated seven different temperatures, ranged from 21 °C to 48 °C, and found that 41 °C was the optimum temperature for short-term methane production. Mata-Alvares and Martina-Verdure (1986) reported the optimum temperature to be between 34 °C and 38 °C, with or without leachate recirculation. Blakey et al. (1997) reported that temperature may be an important factor affecting the methane content of LFG. Operation of landfills under optimum temperatures will result in faster rates of gas production and refuse stabilization. In addition, the transition from the acetogenic to the methanogenic phase can be shortened when landfill is operated under warmer weather. Robinson (2007) summarized that the transition period from the acetogenic to the methanogenic phase of the landfills in temperate countries was double or three times that of the landfills in warmer climates. High methane production and a rapid transition from acidogenesis to methanogenesis can reduce the content of VFAs in leachate, rendering low BOD and BOD/COD ratios.

The actual landfill temperature may vary widely within one landfill. In Nordic conditions, landfill winter temperatures of 5 - 35 °C and summer temperatures of 10 - 45 °C have been measured. The landfill temperature is affected by the size and height of the landfill, climatic conditions and landfilling operations, which determine the circumstances in which microbial decomposition occurs. Understanding the impact of temperature on landfill emissions, especially landfill leachate, is significant for the improvement of long-term landfill management strategies in order to minimize landfill emissions, accelerate waste stabilization and shorten the landfill aftercare period.

The temperature impact on microbial growth rate in anaerobic digestion is usually in the range of 5 - 7% / °C based on data given in Henze et al. (2002) and Metcalf & Eddy (2003). In a more detailed study Siegrist et al. (2002) have shown that from the biodegradation steps hydrolysis has lower temperature sensitivity than the methanogenic step and is around 2 - 3% / °C in the mesophilic and thermophilic ranges. In the work of Reichel et al. (2007) an equation has been used, which clearly indicates an over 10% increase per 1 °C in the range 20 - 35 °C. However, this value is connected to one simulator reactor with a single step temperature increase and does not reflect a long-term adaptation to temperatures near 20 °C. Hydrolysis was the rate limiting step (Vavilin et al. 2003) when the temperatures in landfill reach 55 °C to 66 °C (Berge et al., 2005). It is inhibitory to nitrification because pure Nitrosomonas cultures have a thermal death point between 54 °C and 58 °C (Willers et al., 1998).

In most of the studies referred to, a constant temperature was applied and the impact of different temperature ranges on long-term emissions of landfilled waste has been addressed experimentally in only a few cases, indicating the need for additional studies.

2.3.6 Landfill L/S ratio's impacts on leachate emission

The main premise of bioreactor landfills is the introduction of moisture into the waste through recirculation of leachate or liquids, which beneficially enhances the decomposition rate (McBean et al., 2007). Measuring the amount of water (in litres) passing through a given waste mass (given in kg dry matter of waste), commonly known as the L/S ratio, allows relating the LSR results to full-scale landfills. This L/S ratio is also used to determine the remaining pollution potential of MSW in landfills and, thus, to estimate the time that would be needed to reach the threshold concentration level on the full landfill scale (Fellner et al., 2009).

Fellner et al. (2009) investigated leachate emission variations with L/S ratios in terms of chloride (Cl), nitrogen and TOC at laboratory LSRs and a landfill. Based on tracer experiments, it can be discerned that in laboratory-scale experiments around 40% of pore water participates in advective solute transport, whereas this fraction amounts to less than 0.2% in the investigated full-scale landfill. At a field-scale, and after 16 years of landfilling, the L/S ratio reached 0.60. In a total discharge of 360±50 Cl⁻ mg/kg dry matter was observed, whereas more than double that amount had been discharged from the laboratory reactors (840 mg/kg dry matter) at the same L/S ratio. At the landfill, the chloride load emitted does not exceed 500 mg/kg dry matter at the L/S ratio of 2 (which corresponds to 50 years of landfilling). Assuming no change in water flow conditions, this implies that over 70% of soluble chloride remains inside the landfill. Comparing the Cl⁻ discharge between laboratory LSRs and landfills, the Cl⁻ load remaining in the waste body is larger in landfill. The nitrogen releases at both laboratory LSRs and landfills are comparable. When the L/S ratio reaches 0.60, around 400 mg N/kg dry matter has been emitted from landfill and reactors; this corresponds to less than 10% of the initial nitrogen content (4 - 6 g N/kg dry matter). Results observed at the landfill indicate, however, that future nitrogen loads (L/S > 0.6) will be smaller compared to LSR experiments.

Among the studies referred to, the overall understandings of the L/S ratio's impact within a range higher than 4.0 have been addressed in few cases, indicating the need for further studies.

2.4 Leachate treatment

2.4.1 Leachate treatment alternatives

The major potential environmental impact related to landfill leachate is the pollution of groundwater and surface water (Lema et al., 1988; Manfredi and Christensen, 2009b). In order to reduce the potential risk of damage to groundwater and surface water, leachate should be disposed of before being discharged to receiving water bodies.

In the technical system of leachate disposal, there are three major alternatives for the treatment of landfill leachates, including leachate channelling (e.g. combined treatment with municipal wastewater and recirculation), on-site biological treatment (i.e. aerobic treatment and anaerobic treatment) and on-site chemical/physical treatment (e.g. membrane filtration, chemical precipitation, chemical oxidation, adsorption onto activated carbon, reverse osmosis and ammonia stripping).

If a nearby sewer system is available, and the municipal wastewater treatment plant (WWTP) has adequate capacity, it is popular to treat the leachate at the existing public facility; sometimes the leachate requires on-site pretreatment, depending on its characteristics and the local indirect discharge regulations. The main difficulties are posed by the high concentrations of organic and inorganic components contributed by young and old waste respectively. According to the experience of Finnish wastewater treatment plants, when leachate makes up less than 5% of the total sewage plant input and the leachate COD concentration is 10 g/L or less, then joint treatment is acceptable. Otherwise on-site pretreatment is required before leachate is discharged into the sewer system and the hydraulic retention time (HRT) of the sewage plant should be increased.

As discussed in Section 2.2.1, recirculation is one of the least expensive options available, allowing the achievement of a considerable reduction of both the concentration of organic matter and the volume of leachate. Apart from reducing the leachate strength, biodegradation of organic material in landfill is usually stimulated when the water content reaches up to 50 - 70% due to leachate recirculation (Lema et al., 1988; Jiang et al., 2007).

High concentrations of COD and ammonium have led to the application of biological treatment; various forms of anaerobic treatment have become quite common, at least as a first stage treatment for organic matter in leachate, usually followed by a subsequent aerobic treatment to

further remove ammonium as well as organic pollutants (Kabdaşli et al., 2008). Combined biological aerobic and anaerobic processes can serve to treat organic material more completely and favour several nitrogen transformation and removal processes, including nitrification and denitrification, resulting in the complete in situ removal of nitrogen from landfills.

Relevant chemical/physical methods include membrane filtration, advanced oxidation, activated carbon sorption, ozonation, ultrasound and reverse osmosis. The chemical/physical treatment is effective immediately from start-up, which is easily automated and is insensitive to temperature changes. In most cases, simple materials and a plant are required; but the large quantities of waste sludge generated by the addition of flocculants and the high running cost of the plant and chemicals limit the application of these methods in some cases. So, sometimes chemical/physical methods (e.g. granular activated carbon (GAC) filtration) are only used for the pre- or post-treatment of leachate, to complement biological degradation techniques. In this role, they are especially useful in the treatment of leachates from old landfills and for the elimination of specific pollutants (e.g. humic and fulvic acids) (Lema et al., 1988).

The selection of a particular treatment process highly depends on the quality and strength of the leachate. Generally leachates from the acidic phase are more suitable for biological systems and physical-chemical systems are better for old leachates; but even biological treatment has proved suitable for nitrogen removal, which is the important issue in regard to old landfill leachate. However, the design of a general strategy for the treatment of leachates is hindered by their great diversity, which results in the techniques successfully developed for one site not necessarily being applicable elsewhere.

Treatment methods that work well at a young landfill are likely to become progressively less effective as the landfill ages (Lema et al., 1988; El-Fadel et al., 1997a; Haarstad and Mæhlum, 1999). When treating young (biodegradable) leachate, biological techniques can effectively remove COD, NH₄–N and heavy metals. However, when treating stabilized (low biodegradable) leachate, biological treatment may not be able to meet the discharge limits for organic matter due to the recalcitrant characteristics of organic carbon in the leachate. In recent years, physico-chemical treatments have been found to be suitable not only for the removal of slowly biodegradable substances from stabilized leachate, but also as a refining step for biologically treated leachate. Prior to discharge, additional effluent refinement using physico-chemical treatments, such as chemical precipitation, advanced oxidation, activated carbon adsorption and ion exchange, can be carried out on-site (Kurniawan et al., 2006).

2.4.2 Nitrogen removal processes

A special feature of landfills is their rather low temperature in winter and a lower biodegradation rate of waste. This affects the leachate quality and its treatment. With a low degradation rate, the acid phase can continue for 5 to 10 years or more, which means an elevated COD concentration and a high COD/N ratio in the leachate for this period (Pelkonen et al., 1999). In the EU countries the problem of leachate treatment has existed for some time now, but a universal solution has not been found. The leachates are a mixture of high concentration organic and inorganic contaminants including humic acids, ammonium-nitrogen, heavy metals, xenobiotics and inorganic salts. Taking into account the toxicity of ammonium-nitrogen in leachate, major concerns have been expressed about the treatment of it. There are two strategies with respect to ammonium removal: in-situ treatment and ex-situ treatment. Currently, ammonium treatment is primarily performed via biological co-treatment at municipal WWTPs or via on-site treatment using biological nitrification/denitrification and physico-chemical processes. Biological nitrogen removal from leachate is a challenging task because of the high variation of leachate characteristics and the usually insufficient amount of readily biodegradable organics for denitrification (expressed as the carbon to nitrogen ratio - the C/N ratio). The availability of readily biodegradable organics usually limits the efficiency of biological nitrogen removal, particularly in landfills in the methane phase, resulting in low BOD. Consequently, external carbon addition is required in many landfill leachate treatments (e.g. Pelkonen et al. 2000; Yalmaz and Oztürk, 2001, 2003).

The biological method of nitrification/denitrification is probably the most efficient and cheapest process to eliminate nitrogen from leachate (Abufayed and Schroeder, 1986). Denitrification is a vital step in biological nitrogen removal. In the process, organic and other reducing species or agents are utilized as electron donors by denitrifying bacteria, nitrate and nitrite are reduced finally to nitrogen gas. Denitrification can be carried out either in cooperation with nitrification or independently, in municipal and industrial wastewater treatment. The key to successful independent denitrification is to maintain high biological activity and concentrated activated sludge in the reactor (Peng et al., 2004).

Nitrification/Denitrification

The mechanism of the three major biological processes directly involved with biological nitrogen removal in wastewater treatment can be expressed as:

25

$$NH_{4}^{+} + 1.5O_{2} \rightarrow NO_{2}^{-} + 2H^{+} + H_{2}O \quad [\Delta G_{0} = -275 \text{ kJ/mol}] \quad (1)$$

$$NO_{2}^{-} + 0.5O_{2} \rightarrow NO_{3}^{-} \qquad [\Delta G_{0} = -75 \text{ kJ/mol}] \quad (2)$$

$$Overall: NH_{4}^{+} + 2O_{2} \rightarrow NO_{3}^{-} + 2H^{+} + H_{2}O \quad [\Delta G_{0} = -350 \text{ kJ/mol}] \quad (3)$$

Ammonium is oxidized to nitrate in aerobic conditions by two groups of chemo-lithotrophic bacteria that operate in sequence. The first group of bacteria in this process of nitrification, represented principally by members of the genus *Nitrosomonas* [Eq. (1)], oxidize ammonium to nitrite, which is then further oxidized to nitrate by the second group, usually represented by members of the genus *Nitrobacter* [Eq. (2)].

Denitrification is the second step in the removal of nitrogen by the nitrification/denitrification process. This is a process by which nitrate functions as an acceptor of reducing equivalents and de-assimilates to nitrogen gas:

 $NO_3^- \rightarrow NO_2^- \rightarrow NO \rightarrow N_2O \rightarrow N_2(4)$

The most important issue concerning N removal is to ensure an appropriate C/N ratio. In the activated sludge process, a biodegradable COD/N ratio of around 9 or higher was required in a pilot-scale process to achieve 70 - 90% TN removal (Pelkonen et al. 2000), and in a full-scale realization at a landfill, 55% TN removal was achieved with a biodegradable COD/N ratio of 4.7 (Pelkonen et al. 2000). Meanwhile, Chang (1993) gives a biodegradable COD/N ratio of 4.5 as sufficient in an activated sludge process. The biological process is especially efficient in treatment of young landfill leachates that are rich in VFA. For instance, complete nitrogen removal was obtained during treatment of leachate by means of simultaneous denitrification/metanogenesis in an anaerobic reactor with nitrification (Wiszniowski et al., 2004). On the other hand, when treating leachates characterized by a high level of ammonium and low levels of biodegradable organics, a supplementary source of organic carbon is needed (

and Dzombak, 1991; Ilies and Mavinic, 2001). Also, the development of a control strategy and system to optimize the C/N ratio is worth considering.

In addition, some other biological nitrogen-removal techniques have occurred in the ex-situ treatment of ammonium in leachate; such as partial nitrification and nitrite denitrification, and Anammox.

Partial nitrification and nitrite denitrification

Instead of using full nitrification/denitrification, partial nitrification and nitrite denitrification is one noticeable new strategy, especially when leachate with a high ammonium concentration or low C/N ratios is treated. The main micro-organisms responsible for developing nitrification are the ammonium-oxidizing bacteria and nitrite-oxidizing bacteria. Then, in denitrification, facultative heterotrophic micro-organisms use nitrate and nitrite as oxidants of organic matter to obtain energy and carbon. Because nitrite is the intermediary compound of both ammonium nitrification and nitrate denitrification, it makes it possible to partially nitrify ammonium to nitrite and then denitrify directly from nitrite to nitrogen. Partial nitrification and nitrite denitrification is an approach to reduce the activity of nitrite-oxidizing bacteria and keep up the activity of the ammonium-oxidizing micro-organisms. Previous studies indicate that partial nitrification and nitrite denitrification can reduce oxygen requirements during nitrification and organics requirements during denitrification, as well as the operational costs (e.g. Bae et al. 1997; Fux and Siegrist, 2004; Fux et al., 2006; Spagni and Marsili-Libelli, 2008).

By controlling operational factors like temperature, dissolved oxygen (DO) and pH, it is possible to facilitate ammonium oxidizing and retard nitrite oxidizing, which could restrain the oxygen and organics consumption, and then adjust the applied effective C/N ratio during the biological processes. The temperature has an impact on the activation and sensitivities of both ammonium-oxidizing bacteria and nitrite-oxidizing bacteria. With pure cultures, Grunditz and Dalhammar (2001) showed that the optimum temperature was 35 °C for ammonium-oxidizing bacteria and 38 °C for nitrite-oxidizing bacteria.

A low concentration of DO contributes to the activation of ammonium-oxidizing bacteria and then ensures nitrite accumulation, since the half-saturation coefficients of DO for ammonium-oxidizing bacteria and nitrite-oxidizing bacteria are 0.2 - 0.4 mg/L and 1.2 - 1.5 mg/L, respectively (Peng et al., 2004; Picioreanu et al., 1997). The control of operational DO is found to be critical in affecting the activity of nitrite-oxidizing bacteria and to achieve partial nitrification (Garrido et al. 1997; Bernet et al., 2001; Ruiz et al., 2003). The pH value is sensitive to partial nitrification and nitrite denitrification since it can influence the concentration of free ammonia, free nitrous acid and free hydroxylamine, which are the inhibitors for nitrification and denitrification. Some other operational factors, such as the process scheme or even feeding modes, can affect the availability of degradable organics (as an electron donor) and nitrate or nitrite (as an electron acceptor) in denitrification; in other words, the applied C/N ratio in biological nitrogen removal. Indeed, observations by, for example, Andreottola et al. (2001), Kim et al. (2004) and Puig et al. (2004) suggest the positive impacts of optimizing the biological process scheme and feeding mode on the efficiency of nitrogen removal.

The conditions of inhibiting nitrite oxidation and optimizing the use of readily biodegradable organics are not yet completely understood. It is thought that the permanent inhibition of nitrite oxidation is difficult to maintain in the activated sludge process (Fux and Siegrist 2004); and accordingly the consumption of readily biodegradable organics during nitrification and denitrification is hard to control. Therefore, more tentative work is required before there is a more comprehensive understanding.

Anammox

The anaerobic ammonium oxidation process is a novel, promising, low-cost alternative to conventional denitrification systems (van de Graaf et al., 1996; Strous et al., 1997). The process deals with the conversion of ammonium to nitrogen gas, with nitrite as an electron acceptor, according to the following reaction:

$NH_4^+ + NO_2^- \rightarrow N_2 + 2H_2O$ [$\Delta G_0 = -357 \text{ kJ/mol}$]

The physiology of the anaerobic ammonium oxidizer 'Candidatus Brocadia anammoxidans' was studied in detail. It has a very high affinity for the substrates of ammonium and nitrite. The process is reversibly inhibited by oxygen and irreversibly by nitrite (at concentrations superior to 70 mg N/L for several days) and phosphate (Hellinga et al., 1998). 'Candidatus Kuenenia stuttgartiensis' has a higher tolerance to nitrite (180 mg N/L) and phosphate (600 mg P/L) (Egli et al., 2001). Both bacteria have a similar optimum temperature (37 °C) and pH (8).

The application of the Anammox process to the treatment of high ammonium-nitrogen leachate is particularly promising. It leads to potential savings of up to 60% in oxygen generation and 100% in external carbon, along with significantly reducing the sludge generation and the net emission of carbon dioxide (CO_2) (van Dongen et al., 2001). However, one challenge is how to accelerate the slow growth rate of the bacteria and slow rate of nitrogen removal from these systems (Jetten et al., 2001). Compared with conventional nitrification and denitrification, partial nitritation/Anammox doesn't produce nitrous oxide (N_2O) and CO_2 production and requires lower oxygen demand. Also, no external organic carbon source is needed, resulting in lower operational costs (Fux et al., 2004).

Physico-chemical ammonium treatment

With respect to physico-chemical ammonium treatment, it is found that ammonium stripping and precipitation respectively achieved 94% and 98% NH₄–N removal, with initial NH₄–N concentrations of 3260 mg/L and 5618 mg/L (Kurniawan et al., 2006); but the essential issues to

limit the application include operational costs, leachate strength and the flue gas treatment (Eden, 2000).

The additional costs associated with ex-situ treatment of ammonium have made in-situ removal techniques attractive alternatives. In-situ ammonium treatment is aimed at purifying leachate first and then recirculating leachate back to the landfilled waste, which refers to the discussion in the previous section. Leachate recirculation is an effective way to enhance waste biodegradation and results in a series of positive impacts on the landfill operation, including increased methane production and waste settlement. However, it is not effective in reducing the ammonium-nitrogen concentration; on the contrary, it increases the NH₄-N concentration to some extent. A few in-situ, or partially in-situ, studies have been conducted; however, the data required to enable the adequate implementation of such processes at field-scale bioreactor landfills are lacking.

2.4.3 Lab-scale and technical scale leachate treatment applications

Suspended processes have been applied in full scale to nitrification and denitrification of landfill leachate (Kettunen et al., 1997). Recently, new methods, such as suspended carrier biofilm processes, have also been studied for leachate nitrification, even at low temperatures $5 - 10^{\circ}$ C (Hoilijoki et al., 2000). These processes are reliable, but they normally require major investment, whereas there is also an evident need for low cost and low maintenance systems. Moreover, the treatment processes used should also function in colder climates. An overview of the studies is summarized in Table 4 and the details are discussed hereunder.

Application	Studies
Conventional activated sludge process	Toettrup et al., 1994; Zeghal et al., 1995; Zeghal and
via nitrification/denitrification	Sibony,1996; Kalyuzhnyi et al., 1999; Puznava et al., 2001; Jokela et al., 2002b; Ruiz et al., 2006
Sequencing batch reactors	Mace and Mata-Alvarez, 2002; Teichgräber et al., 2001; Steven et al., 2004; Kim et al., 2004; Pochana and Keller, 1999
Other nitrogen removal processes	Third et al., 2005; van der Star et al., 2007

Table 4 Overview of the studies on landfill leachate treatment

Compact nitrogen removal processes, such as submerged biological aerated filters, have been widely applied on full-scale plants in the last 15 years. The efficiency of this biofilter system

has been widely demonstrated on full scale plants (Zeghal and Sibony, 1996). It started with a down-flow biological aerated filter for carbon removal in the early 80s and, later, up-flow biofilters were developed and used in different configurations for nitrogen and carbon removal, such as secondary nitrification/denitrification in one cell (Toettrup et al., 1994) or tertiary denitrification (Zeghal et al., 1995). Puznava et al. (2001) showed that the biofilter can easily reach nearly 100% ammonium elimination efficiency thanks to a real time aeration control based on online analyzers. The denitrification removal efficiency was about 65 to 75% during all experimental periods.

Kalyuzhnyi et al. (1999) reported that application of an aerobic/anoxic biofilter as the sole polishing step at 10 - 20 °C was acceptable for the elimination of biodegradable COD and nitrogen from the anaerobic effluents approaching the current national limits for direct discharge of treated wastewater.

Jokela et al. (2002b) showed that nitrogen can be removed effectively from municipal landfill leachate by using a nitrifying up-flow biofilter, with waste material as a filter medium, combined with subsequent denitrification of the nitrified leachate in the landfill body. Crushed waste brick can be used as a carrier material in up-flow filters for nitrification of municipal landfill leachate with loading rates of 100 - 130 and 50mg NH_4 -N/L/d at 25 °C and 5 °C, respectively. The nitrified leachate recirculated to landfill body can be over 3.8 g of total oxidized nitrogen (TON)/ton total solids (TS)/day without any adverse effect on the methanogenesis of waste.

Ruiz et al. (2006) also showed that nitrification-denitrification via nitrite may be applied to existing treatment installations with no extra investment costs, which has an important advantage over other new nitrogen removal technologies such as Sharon, Anammox or completely autotrophic nitrogen-removal over nitrite (CANON) processes.

In recent years, the use of sequencing batch reactors (SBRs) in the biological treatment of wastewaters has been widely extended from lab-scale studies to real-scale leachate treatment plants (Mace and Mata-Alvarez, 2002). While lab-scale SBRs have been used for research on carbon and nutrient removal, and the development of urban/industrial wastewater biodegradability assays, real plant applications are still mainly focused on carbon removal. Nevertheless, when operating real plant SBRs the efficiency of nitrogen removal sometimes turns out to be better than the legally required effluent limits (Teichgräber et al., 2001).

Steven et al. (2004) proposed that an on-site wastewater treatment unit, with an upstream supplemental treatment tank, a downstream combination flow splitter, sump and pump box, and a recirculation system, has the potential to substantially remove carbon and nitrogen from wastewater. Though the process was complex, the removal of nitrogen was good.

Kim et al. (2004) and Pochana and Keller (1999) investigated some important parameters such as DO, pH, oxidation reduction potential (ORP), hydraulic retention time (HRT), the C/N affecting nitrogen removal in a SBR. The results indicated that both the ORP and pH could be control parameters for complete denitrification; the control point on the pH profile was not clear using the pulsed pattern of swine waste addition for investigation of denitrification, and with the more sharply changing values of dpH/dt on the control profiles for detection of nitrification. It was suggested that ORP and pH should be chosen as denitrification and nitrification control parameters, respectively.

The CANON process is an innovative, sustainable nitrogen-removal technology for treatment of wastewater containing high concentrations of ammonium nitrogen. In the study by Third et al. (2005), they investigated the enrichment of anammox bacteria from activated sludge and its application in the CANON process on lab-scale. An anammox culture capable of removing 0.6 kgN/m³/d was enriched for 14 weeks in a sequencing batch reactor.

In 2001, the first full-scale anammox reactor in the world was started in Rotterdam (the Netherlands). The reactor was scaled-up directly from laboratory-scale to full-scale and treats up to 750 kg-N/d. In the initial phase of the start-up, anammox conversions could not be identified by traditional methods, but a quantitative polymerase chain reaction (PCR) proved to be a reliable indicator for growth of the anammox population, indicating an anammox doubling time of 10 - 12 days. Reactors with a high specific surface area, like the granular sludge reactor employed in Rotterdam, provide the highest volumetric loading rates. Mass transfer of nitrite into the biofilm limits the conversion of reactor types that have a lower specific surface area (van der Star et al., 2007).

Some of the studies have indicated that it is feasible, from a technical point of view, to find some specific leachate treatment method to improve the leachate management strategy, e.g. the performance of leachate recirculation etc.

2.5 Landfill leachate management

The EU Landfill Directive regulates that landfill aftercare ends only when no activities are required to ensure that no adverse effects on the environment will result from the closed landfill (EU, 1999). The L/S ratio is the governing measure used to explain the course of landfill stabilization, but temperature is also of importance. The results of Heyer's study (2003) show that nitrogen seems to stay longest in the system, for more than 200 years under the conventional way of management; thus, it determines the length of the aftercare period. For this reason, landfill leachate management alternatives that allow the nitrogen problem to be managed in a shorter time and in a more sustainable manner are of great interest. With very long aftercare periods, the management of technical systems will be difficult and the total costs substantial.

Leachate recirculation is an effective way to enhance L/S ratios and results in a series of positive impacts on the landfill operation. Potential leachate recirculation volumes significantly vary depending on waste characteristics, weather conditions, waste compaction and the degradation phase. The optimum volume of leachate recirculation for a specific landfill should be determined by lab-scale and/or pilot-scale reactors prior to field-scale operation. Moreover, the use of leachate recirculation to enhance landfill stabilization is not straightforward and may needs to be supplemented with other enhancement methods including waste shredding, pH adjustment, nutrient addition and temperature management.

Leachate management and treatment costs form the biggest part of operational costs (Heyer et al., 2005) and are therefore of interest. However, more research is needed to look at leachate treatment alternatives and their costs throughout the aftercare period until the stabilization and leachate emission criteria have been met.

3 Materials and Methods

3.1 Landfill

Ämmässuo landfill, as the largest landfill in the Nordic countries, was selected as the test landfill in this study. Around 1.4 million tonnes of municipal waste is landfilled annually in Finland. One third of this amount is disposed of in this landfill (Ministry of the Environment, 2002; Statistics Finland, 2005). The old area of the test landfill covers an area of 52 ha and approximately 10 million tonnes of waste was landfilled in the period 1987 – 2007. The household waste collected in the metropolitan area of Helsinki (with around 1 million inhabitants) constituted the major waste input to the landfill. In 2004, it was estimated that 74% of the landfilled waste was municipal waste; the rest of the received waste was industrial reject or contaminated soil. Regionally, source-separated biowaste collection was adopted since 1996, which dramatically decreased the amount of biowaste landfilled.

As a typical Nordic climate, the highest atmospheric temperature (Figure 3, YTV, 2010) in the test landfill is 15 – 20 °C, in July and August. The coldest season is in January and February, with a temperature of -5 °C. The change in atmospheric temperature shows obvious impacts on the landfill temperatures, especially in the shallow layers. In Figure 4, the selected sample temperature measuring point is close to the middle of the old landfilling area. When the landfill height is less than 5 m, the landfill temperature is in the range of 0 - 25 °C, which is highly variable with the climate and seasons. Then, when the landfill height is greater than 10 m, the temperature is 25 - 40 °C; and when the height is 5 - 10 m, the temperature is 15 - 25 °C. This trend changed in the winter of 2008 with the progress of the final cover construction and the appearance of a thick snow layer. The landfill temperatures increased dramatically; the temperature in the shallow layer was over 20 °C as was hoped. It increased quickly to 30 °C at a depth of 5 m, and to 40 °C at a depth of 10 m. This change happened at all three temperature measuring points with an increase of landfill temperatures; however, at the edge area of the landfill, the increase in temperature was not so clear as in the middle area. It was found that even at the edge area, a stable landfill temperature of 20 °C could be observed in the shallow layer and an average landfill temperature over 25 °C was achieved.



Figure 3 Atmospheric temperatures in the test landfill



Figure 4 A sample of landfill temperatures in the test landfill, 2003 - 2004

The mean rainfall in southern and central Finland is between 600 and 700 mm; but, in the coastal areas, the rainfall is slightly lower. In northern Finland, where about half of precipitation falls as snow, the annual rainfall is about 450 - 600 mm. (The lowest annual rainfall may be 200 to 300 mm and the highest annual rainfall 700 mm in northern Finland and 900 to 1100 mm elsewhere.) The typical precipitation in the test landfill area is illustrated in Figure 5 (YTV, 2010). The annual variation in rainfall shows that the least rain falls in March. The estimated annual precipitation for the test landfill is 700 mm, which is rather representative for southern

Finland. Moreover, the infiltration rate of the test landfill area was estimated to be 45% before the autumn of 2008, as most of areas were covered by vegetation. Then, the infiltration rate decreased to 10% or even lower since the membrane final cover layer was laid in most of the areas. For common Finnish landfills, the infiltration rate is estimated to be 20 - 25%.



Figure 5 Precipitation in the test landfill area

The test landfill has introduced measures to control LFG since 1996. The LFG collection system consists of 220 gas wells, four pumping stations and seven regulation stations (Kouvo, 2005). With the improvement of the gas collection system and the final cover construction added in 2006, about 6000 m³ of gas per hour was collected (corresponding to a collection efficiency of approximately 75%). From 2004, around 75% of the collected LFG is utilized in a district heating boiler through a gas transfer pipe line and the rest is still flared in-situ. The closed area of the test landfill has operated under the current regulation, but does not have – as is the case for most old landfills – a full monitoring record on emissions caused by gas and leachate (Niskanen et al., 2009).

In this study, all waste and leachate samples were taken from the closed area of the test landfill at different periods. Moreover, in the leachate management scenario part of this study, this test landfill is used as a major reference of big landfill.

3.2 Waste

The total amount of landfilled waste in the test landfill is about 10 million tons with an annual rate of around half million tons. 60 - 80% of the landfilled waste is municipal waste. After the

1990s, with source-separation (of paper and cardboard, biowaste, glass, and metals) collection being adopted gradually, only the residual mixed waste was landfilled. The daily waste load was monitored firstly, and then the waste was compacted with a 50-ton landfill compactor in horizontal layers with soils or composted sewage sludge as a daily cover. The mean characteristics (\pm standard deviations) of landfilled waste are summarized in Table 5 (Sormunen et al., 2008). The waste density measured in landfill simulators is 410 kg waste (dry)/m³.

Table 5 Mean characteristics (\pm standard deviations) of landfilled waste in the test landfill

Waste depth,	TS, %	VS_{WET} , %	VS/TS	TKN (kg/ ton dry	Methane potential,
m				matter)	m ³ /ton TS
5 - 13	57±10	35±7	64±14	4.6± 2.8	47±55
15 - 21	52±11	33±9	65±15	3.5±2.1	68±61
27 - 31	50±10	27±1	55±11	2.4± 1.4	21±25
Average	54±10	33±8	63±14	3.9±1.5	50±55

Source: (Sormunen et al., 2008)

3.3 Leachate

As a big-scale landfill, the test landfill generates large quantities of high-strength leachate. The leachate is collected with the bottom liner system and it is led to an equalizing basin. The size of the equalizing basin for landfill leachate is 16 000 m³. The mean characteristics of landfill leachate are summarized in Table 6 (YTV, 2010).

In addition, an equalizing basin for the waters from the recycling field, e.g. the composting plant, is 3000 m³. Including the surface water, all types of wastewater are mixed in the equalizing basin and then pumped to a municipal wastewater treatment plant. The mixture dilutes the leachate and lowers the NH₄-N strength of wastewater but results in a larger wastewater volume and higher loadings. In the year 2009, the amount of wastewater was about 449 000 m³, with the average concentrations of 780 mg BOD₇/L, 2100 mg COD/L, 360 mg NH₄-N/L and 565 mg Cl/L (YTV, 2010). Since the treatment fee is derived from the quality of the leachate, the cost of the current leachate management is considerable; the high total nitrogen concentration of leachate accounts for 65% of the total management cost.

Table 6 Mean characteristics of landfill leachate in the test landfill

Source: (YTV, 2010)

	NH ₄ -N,	BOD ₇ ,	BOD ₂₈ ,	COD,
	mg/L	mg/L	mg/L	mg/L
Average	880	330	600	1830
concentrations				

Some leachate recirculation tests were done between the years 2003 and 2004. A full-scale recirculation system was started when the landfill closure work was completed with a compacted multilayer surface in 2011.

3.4 Landfill simulators (I, II)

Seven LSRs, run with different temperatures and L/S ratios, were used in this study. The longterm landfill leachate and LFG emission performance were systematically simulated, which gave a comprehensive picture about the waste biodegradation process and its affect on the longterm landfill emission potential and landfill aftercare period.

The LSRs used in this study were cylindrical reactors with an effective volume of 130 L, a height of 100 cm and a diameter of 42 cm. A schematic diagram of the LSRs is shown in Figure 6. The LSRs were hermetically sealed to maintain anaerobic conditions and were kept at a constant temperature with the aid of electrical thermostats. From the top gas pipe of the LSRs, LFG was collected to analyse its volume and composition. In the bottom section of the LSRs, leachate was collected and sampled. Moreover, each LSR was connected with a bottom pump to recirculate the leachate every workday. A top feedwater port was used to add deoxygenated tap water.



Figure 6 Schematic diagram of a LSR

A sieve plate was installed in the LSRs to support the MSW filling space. The MSW samples were taken from the test landfill. Wastes were excavated from different depths (namely 2, 5, 10, 17, 25 and 30 m) in order to represent the universal composition of waste in the landfill. The age of the waste samples was estimated to be from 1 to 9 years and the weighted average was around 3.5 years. These (wet) waste samples contained around 48.5% dry matter, of which 45% was volatile solids (VS). The mixed wastes were on average divided into seven LSRs: R1 - R7. The average waste density was 850 kg waste (wet)/m³.

In the start-up phase, all seven LSRs were operated at 20 °C. After around a 9-week initial phase, five of them were adjusted to 32 °C (mesophilic), and two reactors were adjusted to 46 °C (thermophilic). The remaining two reactors were kept at 20 °C (close to psychrophilic). Every workday, the leachate was recirculated at a rate of 4.5 or 2.25 L/day. Moreover, some leachate was removed weekly from the LSRs, and the same amount of deoxygenated water was sequentially added to the LSRs. The parameters of these operations are listed in tables 7 and 8.

	Psychrophilic		Thermo	ophilic	Mesophilic		
LSR	R1	R4	R2	R3	R5	R6	R7
Waste load (kg dry waste)	43.7	43.6	43.3	44.4	43.8	43.1	43.9
Waste loading rate (kg dry waste/m ³)	421	422	395	439	408	392	418

Table 7 Thermal regime and waste charged in the LSRs

In this study, the conditions of the LSRs were monitored by measuring the LFG and leachate. All the LFG was collected using aluminium-coated gas bags (Tesseraux, Germany). The LFG volume was continuously measured using a Ritter TG 05/5 drum-type gas meter, and its composition (methane and CO_2) was periodically analysed using an LFG10 LFG analyser. The leachate was sampled every two or three weeks. The temperature, pH and conductivity of all the leachate samples were measured in-situ using the portable meters: WTW pH 323 and WTW LF 320. The redox of some leachate samples was measured using a WTW senTix ORP combination electrode. The NH₄-N, NO₂-N, and NO_X-N of the leachate were determined using flow injection analysis (FIA). These measurements followed the international standard ISO 11732 (ISO International Standard, 2005) and the Finnish standard procedure SFS-EN 13395, ISO 2005 and SFS 1997 (SFS Finnish Standards Association, 1997). The COD_{Cr} was analysed using SFS 5504 1988 (SFS Finnish Standards Association, 1988).

		Psychrophilic		Thermo	ophilic	Mesopl	Mesophilic		
LSRs		R1	R4	R2	R3	R5	R6	R7	
	Period, days								
Temperature, °C	0-63	20	20	20	20	20	20	20	
	63 - 100	20	20	32	32	32	32	32	
	100 - 1400	20	20	46	46	32	32	32	
Recirculation,	0-430	4.5	4.5	4.5	4.5	4.5	4.5	4.5	
L/day	430 - 1400	2.25	2.25	2.25	2.25	2.25	2.25	2.25	
Removal/addition,	0-63	Start-up phase, 2.0 -0.75							
L (water)/week	63 - 810	0.75	0.5	0.75	0.75	0.75	0.75	0.2	
	810 - 1250	1.5	1.0	1.5	1.5	1.5	1.5	0.4	
	1250 - 1400	0.75	1.0	0.75	0.75	0.75	0.75	0.4	
Average applied annual	0-63	Start-up phase, 1.50 – 1.55							
L/S,	63 - 810	0.93	0.61	0.94	0.93	0.93	0.95	0.29	
L(water)/kg (ury waste)	810-1250	1.80	1.19	1.83	1.81	1.81	1.84	0.48	
	1250 - 1400	1.29	1.22	1.29	1.30	1.29	1.31	0.49	
Simulated real landfill	0-63	Start-up	phase	I				1	
time in Finland, years	810	96	67	96	96	95	97	37	
	1250	171	117	172	171	170	173	57	
	1400	207	151	207	209	206	210	71	

Table 8 Operational parameters of the LSRs

¹⁾ related to a big landfill of 25 m height, 700 mm annual precipitation and 25% infiltration – rather typical values in many European countries.

3.5 Biological leachate treatment and cost estimation (III, IV)

With landfill leachate taken from the test landfill, the lab-scale biological leachate treatment processes with a sequential batch reactor were operated for a period of around 800 days. In this study, as a crucial leachate management alternative, the feasibility of an on-site process to treat the leachate (mainly TN) for both direct discharge and indirect discharge purposes was evaluated. More importantly, with a case study of cost estimation based on the test landfill's

conditions, the economic analysis was integrated as an important part of the study to define the applicability of the alternative.

The schematic diagram of the SBR with activated sludge is shown in Figure 7. The SBR process is composed of a cylindrical reactor with an effective volume of 9.5 L. This process was operated at 25 ± 1 °C in a thermostatic room. pH was controlled and tested in the range of 7.8 – 8.2 and a buffer addition pump was used to adjust pH as necessary.



Figure 7 Schematic overview of a sequential batch reactor with activated sludge

A data acquisition and control programme was utilized to control these sequences. Feeding was divided into six steps in a cycle, and a total of around 0.6 L of the influent leachate was fed into the reactor in each cycle. The aeration phase was achieved by injecting compressed air with a flexible spiral diffuser. The compressed air supply level was pre-set, based on the organic matter concentration of the influent. A DO measurement system was used to monitor the real time DO values, which was used as the control indicator of aeration time. If DO values were more than

1.1 mg/L, aeration was interrupted. This limitation was adjusted to 0.6 mg/L, as the external carbon addition was 6 ml/day for the partial nitrification and nitrite denitrification test.

A so-called humidity tank was employed within the aeration system in order to reduce intense evaporation. A magnetic stirring apparatus continually worked at a speed of 30 rpm to maintain homogeneity in the reactor. The other accessory apparatus included a leachate filling pump, a pressure-operated emptying valve and an acetate addition system. 100 g/L CH₃COO⁻ solution was selected as the external carbon addition agent.

The seed sludge used in this process was taken from a Finnish landfill leachate treatment plant and a municipal WWTP. It was incubated within the previous studies (e.g. Wang, 2004) and the mixed-liquor suspended solids (MLSS) were kept at approximately 3400 – 6200 mg/L.

The influent feeding landfill leachate was periodically taken from the test landfill. Many factors, such as climate, season, waste character and landfill age, can affect leachate characteristics. As a multi-area landfill, 60% of the landfill area in the test landfill has been used for over 9 years, but some new areas have also been gradually brought into use in recent years. Hence, the leachate quality varied. The raw leachate presented mean values of 831 mg NH₄-N/L, which ranged from 523 - 1108 mg NH₄-N/L. The BOD₇/NH₄-N (BOD/N) ratio fluctuated in the range of 0.2 - 0.5. This BOD/N range was comparatively low even for the aged landfills (Farquhar, 1989). External carbon was added to adjust the biodegradability of the influent feeding leachate, which presented mean values of 2763 mg COD/L and 618 mg BOD₇/L, respectively, ranging from 1300 - 4280 mg COD/L and 105 - 1490 mg BOD₇/L.

Ten stages (tables 9 and 10) were identified according to the external carbon addition levels (ml 100 g/L CH_3COO^{-}/day). From Stage 1 (external carbon addition level: 0) to Stage 7 (10), the external carbon addition level was increased step by step to protect the adjustability and flexibility of the process, and then it was stepped down to stage 10 (1.5). The aims of this arrangement were to minimize the influences of the variety of the leachate quality and to test the performance of biological nitrogen removal with the broader C/N range.

In summary, the operational parameters and applied influent (e.g. the HRT, the applied C/N ratio, the organic loading rate [OLR] and applied nitrogen loading rate [NLR]) in different stages are listed in Table 10.

			Stag	e 1	Stage	2	Stag	e 3			Stage	4	
External carbon			0		0.7		1.5				3		
Influent leachate No.	Overa	.11	1 2	3	3		3	4	4		5	6	7
Average BOD ₇	327	N	A N	A 239	239) 2	39	390	39	0 3	372	437	315
Average BOD ₂₈	597	N	A N	A 455	455	6 4	55	631	63	1 :	507	727	524
BOD ₇ /BOD ₂₈	0.60	N	A N	A 0.53	0.53	3 0	.53	0.62	0.6	52 0	0.73	0.60	0.60
Average COD	1826	5 18	40 228	33 197	4 197	4 19	974	2161	210	51 2	002	1696	2085
Average NH ₄ -N*	880	9	47 100	01 847	847	8	47	960	96	0 5	562	919	1014
continued		•			-								
		Stage 5	5		Stage 6		Stage	e 7	Stag	ge 8	Stage	9 St	age 10
External carbon		6			8		10	'	5	5	3		1.5
Influent leachate No.	7	8	9	9	10	11	12		13	14	14		14
Average BOD ₇	315	434	355	355	361	451	394	4	321	265	265		265
Average BOD ₂₈	524	749	575	575	563	727	828	8 (637	417	417		417
BOD ₇ /BOD ₂₈	0.60	0.58	0.62	0.62	0.64	0.62	0.4	8 0).50	0.64	0.64		0.64
Average COD	2085	2206	1847	1847	1572	2261	152	3 1	285	1331	1331		1331
Average NH ₄ -N*	1014	1125	1005	1005	841	1088	664	4 0	669	720	720		720

Table 9 Quality of raw influent landfill leachate (Not analysed [NA]) (Units: External carbon, ml/day; others, mg/L)

Parameters	Stage 1	Stage 2	Stage 3	Stage 4	Stage 5
External carbon, ml/day	0	0.7	1.5	3	6
HRT, days	13.8±0.6	15.0±2.0	14.4±1.4	15.8±4.2	13.5±1.5
Operational days	84	18	31	91	159
No. of observations	23	5	8	26	44
Applied C/N	0.3	0.2	0.4	0.7	1.2
OLR, mg BOD/g VSS/d	8±4	6±2	15±1	26±12	50±14
Influent COD, mg/L	2115±163	1977±68	2379±291	2506±220	3157±268
NLR, mg N/g VSS/day	64±10	63±15	69±6	62±12	68±9
Influent NH ₄ -N, mg/L	996±62	916±107	895±41	924±34	920±71
Parameters	Stage 6	Stage 7	Stage 8	Stage 9	Stage 10
External carbon ml/day	0	10	5	3	15
External earbon, mi/day	8	10	5	5	1.0
HRT, days	8 13.2±0.5	12.7±0.4	13.5±0.9	13.4±0.4	13.6±0.2
HRT, days Operational days	8 13.2±0.5 113	10 12.7±0.4 35	13.5±0.9 34	13.4±0.4 28	13.6±0.2 98
HRT, days Operational days No. of observations	8 13.2±0.5 113 32	10 12.7±0.4 35 11	13.5±0.9 34 11	13.4±0.4 28 8	13.6±0.2 98 24
HRT, days Operational days No. of observations Applied C/N	8 13.2±0.5 113 32 1.7	12.7±0.4 35 11 2.8	13.5±0.9 34 11 1.6	13.4±0.4 28 8 1.1	13.6±0.2 98 24 0.7
HRT, days Operational days No. of observations Applied C/N OLR, mg BOD/g VSS/d	8 13.2±0.5 113 32 1.7 64±13	10 12.7±0.4 35 11 2.8 58±3	34 11 1.6 27±5	13.4±0.4 28 8 1.1 18±7	13.6±0.2 98 24 0.7 8±2
HRT, days Operational days No. of observations Applied C/N OLR, mg BOD/g VSS/d Influent COD, mg/L	8 13.2±0.5 113 32 1.7 64±13 3565±394	10 12.7±0.4 35 11 2.8 58±3 3442±171	3 13.5±0.9 34 11 1.6 27±5 2314±171	13.4±0.4 28 8 1.1 18±7 1975±44	13.6±0.2 98 24 0.7 8±2 1601±72
HRT, days Operational days No. of observations Applied C/N OLR, mg BOD/g VSS/d Influent COD, mg/L NLR, mg N/g VSS/day	8 13.2±0.5 113 32 1.7 64±13 3565±394 49±8	10 12.7±0.4 35 11 2.8 58±3 3442±171 31±3	3 13.5±0.9 34 11 1.6 27±5 2314±171 31±3	13.4±0.4 28 8 1.1 18±7 1975±44 34±4	13.6 ± 0.2 98 24 0.7 8\pm2 1601\pm72 37\pm2

Table 10 Operational parameters and influent (average COD, NH_4 -N concentrations-stages) of the sequential batch reactor with activated sludge (mean or mean \pm standard deviation)

Two operational strategies and operational schemes – schemes PA and PB – were studied, which are illustrated in Figure 8. Each 24-hour cycle consisted of six sequences: influent feeding, mixing, an aeration phase, an anoxic phase, a settlement phase and emptying. Figure 8 shows the distribution of the aerobic and anoxic phases, scheduled over six anoxic-aerobic combinations. A filling event (0.25 - 0.4 min) was carried out at the start of each anoxic-aerobic

combination. The filling time was two minutes for each cycle, which is equal to around 0.6 L influent filling. With the same C/N ratio, the impacts of two aeration schemes on DO control and nitrite oxidation were tested. The step-filling strategy was adopted to enhance denitrification, using the easily biodegradable organic matter from the leachate.



Figure 8 Operational cycles of a sequential batch reactor with activated sludge

The influent and effluent were sampled at least twice a week; and the mixed liquor was sampled about once a month. The influent and effluent samples were filtrated with Schleicher & Schuell GF50 filter paper. The temperature, pH and conductivity, NH_4 -N, NO_2 -N and NO_X -N, of all the samples were measured (as shown in Section 3.4) and the PO_4^- of the samples were determined as well, using flow injection analysis (FIA). These measurements followed international and Finnish standard procedures (ISO, 2005 and SFS, 1997, 2004, 2005). Moreover, the BOD₇ (European standard EN 1899-1, 1998), COD_{Cr} , suspended solids (SFS-EN 872, 1996) and total solids (SFS 3008, 1990) of the samples were also periodically detected. The long term BOD_{28} was determined using the WTW OxiTop system at 20 °C.

The results were analysed statistically with the Stat View programme (SAS Institute, 2001). Analysis of variance and a t-test were used to find out the difference of means of two groups. Multiple regression analysis was used to model selected independent variables.

Finally, based on the process developed in this study, the cost-savings from the actual leachate management costs were estimated systematically to identify the possibility of optimizing operational costs. The raw leachate flow rate was assumed as $500 \text{ m}^3/\text{day}$ and TN concentration

as 850 mg/L. Methanol was taken as the external carbon source and its price was set as 365 \in /tonne. The overall leachate management cost estimation includes investment costs, general pretreatment operation and maintenance costs, chemical costs and the leachate treatment fee of the WWTP. The basic approach is calculated for 60% TN removal (investments). When higher than 60% TN removal is targeted, two options are included – one as an extension of the single sludge system with higher carbon needs for the additional (over 60%) TN removal, and another as a post-denitrification (post DN) system with its own sludge (two sludge systems), also for additional TN removal. In this case the carbon need is estimated as 3.7 g COD/g NOx removed and the specific denitrification rate of 5 mg NOx/g VSS/h (an average value measured with batch tests) has been used for the reactor volume estimation.

3.6 Leachate management scenario and strategy model (V)

3.6.1 Forecast modelling for target variables

The cumulative landfill leachate emission was calculated by treating the leachate quality/quantity as a function of temperature and the annual L/S ratio. When the temperature is constant, the relationship between the target components and the L/S ratio can be abstracted as follows:

Leachate:

 $y = ae^{bl}$

LFG:

 $y = al^b + c$

where

y represents the concentration of the target component, e.g. COD, NH₄-N, the methane generation rate and so forth,

l represents the L/S ratio and

a, b, c are the coefficients.

a, *b* and *c* are determined by using linear regression analysis. According to the residual analysis, the simulated range can be reasonably extended based on this model. To simplify the modelling,

the simulation is started from the peak value during the acidic phase and the short initial aerobic phase is omitted.

The L/S ratio in the equation refers to fresh water addition to the system. Or, alternatively, in the case of nitrogen leaching, it can refer to the addition of leachate with full nitrification in the pretreatment step (discussed in more detail later).

With a constant L/S ratio rate, the relationship between the target variables and temperature is estimated via the temperature coefficient, θ :

$$y = y_t \theta^{(T-t)}$$

where

T represents the target temperature [$^{\circ}$ C],

y represents the concentration of the target component, e.g. COD, NH₄-N, the methane generation rate and so forth, at the target temperature,

t represents the reference temperature [°C] and

 y_t represents the concentrations of the target component, e.g. COD, NH₄-N, the methane generation rate and so forth, at the reference temperature.

3.6.2 Forecast modelling for the leachate emission integrator

Based on the emission limit values, with the forecast model it is possible to evaluate the length of time needed for the leachate to meet the direct discharge requirement. Some variables, for example NH_4 -N, will take a longer time to descend to the target level, which means that they will have heavier impacts on the length of landfill aftercare. Hence, the weight coefficient of each target variable is assumed accordingly. The leachate emission integrator *LE* is defined as:

$$LE = \sum_{i} \alpha_{i} \frac{y_{i}}{Y_{i}}$$

where

 y_i represents the concentrations of the target components, i.e. COD, NH₄-N and Cl⁻ (representatives of organic and inorganic components),

 Y_i represents the emission limit values for the target components and

 a_i represents the weighting coefficients for the target components.

When the concentration of a target component meets the emission limit value, the value of $\frac{y_i}{y_i}$

is 1 and the value of $\alpha_i \frac{y_i}{Y_i}$ is α , which is the minimum level.

3.6.3 Leachate management scenarios

The integrators are applied and compared in six different forecast scenarios (Table 11). These landfill scenarios represent various landfill sizes and annual L/S ratios based on the general condition of existing sanitary landfills. The landfill filling period is a 20-year period; no leachate recirculation system will be used before landfill closure. The conversion between the achieved L/S ratio and real landfill time refers to a typical landfill with 200 mm of annual natural infiltration. The selection of the temperature range depends on the landfill size. The annual L/S ratio rate was selected accordingly. For big landfills, an annual L/S ratio rate of 0.013 corresponds to a one-fold infiltration, whereas 0.038 corresponds to a threefold infiltration (extra water 400 mm /year). An annual L/S ratio of 0.076 includes 700 mm/year of fresh water and 300 mm/year recirculation of treated leachate effluent. For medium-sized landfills, an annual L/S ratio rate of 0.034 corresponds to a one-fold infiltration, whereas other infiltration rates are similar to those of big landfills. The use of fresh water, such as surface water collected in the landfill area, is aimed at chloride washout and the recirculation of treated leachate effluent is related especially to nitrogen.

	Recirculation	Water infiltration, mm/year	Annual, L/S	Temperature, °C			
			(L/kg TS/year)				
Big landfill, averag	e waste height: 25	m (area 50 ha, 10 Million tons, 630 kg TS/m ³)					
Scenario A	No	Natural infiltration, 200	0.013	32			
Scenario B	Yes	Natural infiltration, 200 +	0.038	32			
		Fresh water recirculation,					
		400 = 600					
Scenario C	Yes	Natural infiltration, 200 +	0.076	32			
		Fresh water recirculation,					
		700 + on-site treatment					
		process effluent $300 = 1200$					
Medium-sized land	fill, average waste	e height: 10 m (area 25 ha, 1.9 l	Million tons, 580	kg TS/m ³)			
Scenario E	No	Natural infiltration, 200	0.034	20			
Scenario F	Yes	Natural infiltration, 200 +	0.103	20			
		Fresh water recirculation,					
		400 = 600					
Scenario G	Yes	Natural infiltration, 200 +	0.207	20			
		Fresh water recirculation,					
		700 + on-site treatment					
		process effluent $300 = 1200$					

Table 11 The general specifications of the six scenarios

Scenarios B, C, F and G may need some amount of leachate recirculation.

3.6.4 Landfill leachate management strategy model

The above work will produce results that describe the status of the landfill leachate in a comprehensive profile, which is combined with different treatment alternatives for the evaluation of applicable strategies. On-site and off-site treatment alternatives included in this study are shown in Figure 9.

An on-site biological leachate treatment process has been proven to be feasible from both technical and economic points of view in previous studies, and it can be applied before the leachate recirculation or as a pretreatment process before indirect discharge. The biological process (nitrification, partial denitrification) can also be adopted with/without post DN or GAC filtration to meet the direct discharge limit value.

In the unit cost estimation, a 10 - 30 year depreciation time and a 6% interest rate have been used (Wang et al. 2009). The estimated future costs of discharge for a municipal WWTP are based on the current unit costs of leachate components and the flow rate. The water infiltration costs have been estimated at 3900 ϵ /ha/year, including investment and operation (Okereke, 2002). When the six scenarios are compared, the aftercare time period comes to hundreds of years and the total costs are calculated based on zero interest rates.


Figure 9 Leachate treatment options for cost estimation (*advanced treatment needed when effluent COD does not meet the limit; $**\pm 25\%$ unit cost changes used for cost estimation)

3.7 Landfill leachate discharge limits

The EU Landfill Directive stipulates that leachate has to be treated properly before it is discharged to surface waters, but the directive doesn't provide any concrete discharge limit values. These are left to be formulated by the EU Member States. The decision regarding the leachate discharge limit values strongly recommends the strategy of leachate management and landfill aftercare. As examples, some general discharge limit values of the major target components in this study are presented in Table 12. The regulations in European countries may vary from more general regulations to site-specific considerations, which shows the complexity behind the aftercare problem and the regulations for dealing with it (Scharff et al., 2011). In this study it is assumed that the limit values for leachate discharge determining the end of the aftercare period are 200 mg COD/L, 30 mg BOD/L, 70 (70) mg TN (NH₄-N)/L and 100 mg/L for chlorides. These are mainly based on German limit values (Heyer, 2003; Stegmann et al., 2006), as there are no such general limit values given in Finland. Based on some local (sitespecific) regulations in Finland, the German limits are at the same level or stricter. Regarding LFG emission, it is assumed the limit value to be 1 L/kg waste (dry)/year for big landfills; for medium-sized landfills, the emission limit value for aftercare should not exceed 1.5 L/kg waste (dry)/year, based on data given by Heyer et al. (2005) and assuming appropriate waste heights.

Table 12 Major leachate discharge limit values in some countries, mg/L

	Direct	Indirect			
	Germany	France	China	Japan	Australia
COD	200	120	100	90	1500
BOD	20	30	30	60	600
TN (NH ₄ -N)	70 (NR)	30 (NR)	40 (25)	60 (60)	150 (100)

(China Environmental Science Institute, 2007)

4 Overview and discussion of results

In this chapter, the first section is dedicated to lab-scale LSRs, to express the simulated tendency of leachate emissions. The second and third sections are a discussion of on-site leachate treatment solutions regarding biological nitrogen removal and the related cost estimation issues. The last section is to scale up and forecast the long-term leachate emission performance in real landfill and landfill leachate management strategies.

4.1 Leachate emission and biodegradation of waste (I, II)

Seven LSRs were operated over 175 weeks. Among the seven reactors, which ran at different temperatures and with various L/S ratios, two pairs of parallel reactors were operated under the same conditions, which showed similar behaviour results with only minor differences. It indicates that the aim of careful filling and operation of the LSRs was accomplished and that the material in each reactor was very similar. Meanwhile, between the different applied annual L/S, LSRs under the same operational temperature, i.e. R1 (achieved L/S ratio: 4.1) – R4 (2.7) and R5 and 6 (4.1) – R7 (1.3), the main parameter results were very similar (\pm 7%) with the same L/S ratio level. Thus, it can be concluded that the higher annual L/S ratio rate does not seem to bias the results. This verifies the feasibility of shortening the experimental time using a higher annual L/S ratio rate. To simplify the analysis, the average value of the high annual L/S ratio reactors (R1, R5 and R6) were used for the analysis, and the low annual L/S ratio reactors (R4, R7) were used as a reference as needed. Based on assumptions shown in Table 8, the time in high annual L/S ratio reactors to achieve an L/S ratio of 1 was estimated to correspond to an actual landfill period of 47 years.

4.1.1 Leachate emissions

NH₄-N, COD (organics) and chloride (inorganics) are selected as the key parameters of leachate emissions to be discussed, which are summarized in Figure 10.



Figure 10 Variation of leachate NH_4 -N, COD and Cl⁻ as a function of temperature and the L/S ratio

54

Not considering the temperature adjustment phase, the NH_4 -N concentrations decreased from 1100 - 1500 mg/L to less than 200 mg/L, with an L/S ratio of around 4.3; but they are still much higher than the direct or indirect discharge limit value, both in the low and high temperature LSRs. To take a mesophilic LSR as an example, with the same L/S ratio level (e.g. 2), the NH_4 -N concentration measured in this study was around 1.5 times higher than the measurements of Fellner et al. (2009). Also, the L/S ratio levels to reach the estimated NH_4 -N direct discharge limit value were higher than in the studies of Andreas and Bilitewski (1999), Andreas (2000) and Heyer (2003); the higher organic fraction in the sampled waste is considered to be the main reason for this.

Owing to the rapid hydrolysis of the organics of MSW, the COD concentrations increased to 21 000 mg/L, from an initial 15 000 mg/L, within the first six weeks (an L/S ratio lower than 0.26). Then, the COD concentration started to decrease, reaching the level of 4 000 mg/L, as the temperature adjustment phase ended (an L/S ratio of around 0.4), and the level of less than 1 000 mg/L after 140 weeks (L/S ratio= 2.9). The impact of temperature emerged clearly when the L/S ratio increased to 0.55. Higher temperatures obviously impelled the organics' degradation in the LSRs, which resulted in higher leachate COD concentrations in the thermophilic and mesophilic LSRs. These COD concentration levels are still much higher than the limit level of leachate direct (or even indirect) discharge. The psychrophilic LSRs' leachate reached the indirect discharge level of below 1500 mg/L when L/S=1.1. The mesophilic LSRs then achieved it when L/S=1.3; and the thermophilic LSRs with an L/S ratio of around 1.5. When L/S=4.15, the psychrophilic LSRs' leachate COD concentration reached a direct discharge level below 200 mg/L. Although the effect of the temperature was decreasing, the COD concentration of the thermophilic LSR was still far from the limit value. The psychrophilic LSRs' leachate met the limit value for BOD emission when L/S=1.7. The mesophilic LSRs then achieved it when L/S=2.3; and the thermophilic LSRs when L/S=2.8. Therefore, BOD showed better potential for achieving the limit values, which is in accordance with the results Heyer (2003) found in mesophilic simulators.

In week 28 (L/S = 0.65), the psychrophilic LSRs leachate Cl⁻ concentration was 1635 mg/L, which was 85% of the mesophilic (1920 mg/L) LSRs and 84% of the thermophilic (1950 mg/L) LSRs. In week 66 (L/S = 1.30), the chloride concentration decreased to 1170 mg/L, which was 87% and 91% of the mesophilic (1345 mg/L) and thermophilic (1290 mg/L) values, respectively. When L/S = 4.15, the psychrophilic LSRs' leachate was 200 mg Cl⁻/L, which is still much higher than the limit value. Similarly with NH₄-N, the Cl- concentration and emission

potential were bigger (around 2.3 times bigger) than the measurements of Fellner et al. (2009). The difference in the composition of sampled waste may be the main reason.

4.1.2 Cumulative gas production and methane content

In week 28 (L/S = 0.65), thermophilic LSRs generated accumulatively 73 L gas/kg TS, which was 53% more gas than psychrophilic LSRs and 4% more gas than mesophilic LSRs. When L/S=4.15, this value was 110 L gas/kg TS, which was 47% more gas than psychrophilic LSRs and 9% more than mesophilic LSRs. Comparing with the reference levels measured by Sormunen et al. (2008) and by Laner (2011), this amount was higher than the average but still in the range, which is in accordance with the findings of the comparison between field investigations and LSRs by Fellner et al. (2009).

When L/S=4.15, the LFG generation rates at the different temperatures were almost the same: psychrophilic condition, 5.4 L/kg waste (dry)/year; mesophilic condition, 4.9 L/kg waste (dry)/year; and thermophilic condition, 5.5 L/kg waste (dry)/year. All of them are higher than the limit value for aftercare.

The residual VS contents after an L/S ratio higher than 4.35 were 25% in psychrophilic conditions, 28% in mesophilic conditions and 26% in thermophilic conditions. With this L/S ratio, the stable CH₄ contents of the mesophilic LFG was around 56%, which was higher than the thermophilic one (51%) and lower than the psychrophilic one (58%). The generated methane volumes were 187 L CH₄/ kg VS in the psychrophilic LSRs, 289 L CH₄/ kg VS in the mesophilic LSRs, and 288 L CH₄/ kg VS in the thermophilic LSRs. In general, higher temperatures stimulated the waste degradation and methane generation, which is meaningful for LFG utilization. A psychrophilic condition results in lower LFG emission and CH₄ generation, but it seems that it would not extend the waste stabilisation period compared to a thermophilic condition. The gap between the LFG emission amounts at different temperatures is mainly caused by the different LFG emission rates with an L/S ratio below 1. The temperature coefficients of LFG emission are discussed more in Section 4.1.3.

4.1.3 Temperature impacts

The impacts of temperature emerged in all the parameters mentioned. The thermophilic LSRs showed the highest cumulative gas production, gas generation rate, leachate COD and NH_4 -N concentrations, whereas the psychrophilic LSRs resulted in the lowest levels. It is clear that before the landfill closure (e.g. 30 years) all the parameters of the leachate under all the temperatures mentioned are still much higher than the limit values.

The decreasing rates of COD and NH_4 -N concentrations were also temperature-dependent, which means that the decrease of leachate concentration was faster under higher temperatures; however, this impact will decrease when a high L/S ratio is achieved. Meanwhile, the thermophilic LSRs had the highest overall degree of degradation (6% higher than the mesophilic LSRs and 55% higher than the psychrophilic LSRs). After an L/S ratio over 4, the waste degradation degrees under mesophilic and thermophilic conditions were quite close, which was around 1.5 times that found under psychrophilic conditions. Hence, the increase in temperature can accelerate waste degradation and gas generation but cannot decrease the length of the waste stabilisation period since it will extend the waste degradation degree. To considerably decrease the stabilisation period, a higher annual L/S ratio rate was verified to be an effective factor.

The temperature coefficients for different parameters as a function of the L/S ratio are shown in Table 13. The results indicate that the temperature increase showed a rather similar impact on conductivity, chloride and NH_4 -N. It means that the emissions increased synchronously with the increase in temperature, and the rate of increase was approximately the same in psychrophilic, mesophilic and thermophilic conditions.

Higher temperatures obviously impelled the NH₄-N release in the LSRs, which resulted in higher leachate NH_4 -N concentrations in the thermophilic and mesophilic LSRs. The temperature impact on NH₄-N leaching was more obvious at a low L/S ratio; with the increase of the L/S ratio, this phenomenon was weakened. The concentration of NH₄-N showed a slower decline and a long-term consistent tendency in leachate. This suggests that NH₄-N will be of most concern in the long run and more attention should be paid to the effective removal of NH₄-N from leachate.

For the COD emission, as the temperature changed from mesophilic to thermophilic, the value of the COD increased by 1.5%/°C, which was 67% higher than when the temperature changed

from psychrophilic to mesophilic. This confirms that a high temperature is advantageous for waste stabilisation and organics leaching.

For chloride emission, the difference (standard deviation) among the different temperatures decreased to 34 mg/L when L/S=4.15, compared with 104 mg/L when L/S=1.30. The higher temperature activated the waste degradation and chloride emission, but this phenomenon weakened with a L/S ratio above 2.

A higher operational temperature accelerated the gas generation. The difference caused by the temperatures mostly occurred with an L/S ratio lower than 1. This phenomenon faded after the L/S ratio was over 1.53. The highest sensitivity to temperature was related to gas generation (1.6 – 2.8%/°C), which indicates most closely the biological activity. It was found that, as the temperature changed from mesophilic to thermophilic, the value of the LFG emission rate increased 2.8%/°C, which was 1.75-fold of the impact caused by the change from mesophilic to thermophilic conditions. Moreover, the temperature coefficients of the methane generation rate were close to each other.

The temperature coefficient found here differs considerably from the scarce studies of landfill simulations related to temperature; but a temperature coefficient related to hydrolysis can be expected and the findings of this study are close to the value given by Siegrist et al. (2002). Moreover, the LSR results extend experimental data to the lower thermophilic range. The temperature range in this study covers rather well the temperature ranges found in field conditions, shown before and recently by Hanson et al. (2010). As a result, when estimating long-term leachate emission and waste degradation of landfills, one must be cautious and select temperature coefficients carefully, based on representative data. Otherwise misleading emission development may result, which will affect landfill management options in long-term and life cycle inventories.

L/S ratio	Estimated temperatur	re coefficient, θ : C= C	$C_t \times \theta^{(1-t)} *$	
	Chloride, mg/L	NH ₄ -N, mg/L	COD, mg/L	LFG, L/kg TS
0.6	1.016 (20 – 32°C)	1.017 (20 – 32°C)	1.008 (20 – 32°C)	1.034 (20 – 32°C)
	1.006 (32 – 46°C)	1.014 (32 – 46°C)	1.013 (32 – 46°C)	1.017 (32 – 46°C)
1	1.014 (20 – 32°C)	1.014 (20 – 32°C)	1.011 (20 – 32°C)	1.028 (20 – 32°C)
	1.006 (32 – 46°C)	1.012 (32 – 46°C)	1.013 (32 – 46°C)	1.016 (32 – 46°C)
1.5	1.013 (20 – 32°C)	1.015 (20 – 32°C)	1.007 (20 – 32°C)	1.028 (20 – 32°C)
	1.006 (32 – 46°C)	1.012 (32 – 46°C)	1.013 (32 – 46°C)	1.017 (32 – 46°C)
2	1.009 (20 – 32°C)	1.009 (20 – 32°C)	1.009 (20 – 32°C)	1.027 (20 – 32°C)
	1.009 (32 – 46°C)	NH ₄ -N, mg/L 1.017 (20 - 32°C) 1.014 (32 - 46°C) 1.014 (20 - 32°C) 1.012 (32 - 46°C) 1.015 (20 - 32°C) 1.012 (32 - 46°C) 1.015 (20 - 32°C) 1.010 (32 - 46°C) 1.009 (20 - 32°C) 1.010 (32 - 46°C) 1.011 (20 - 32°C) 1.005 (20 - 32°C) 1.016 (32 - 46°C) 1.015 (20 - 32°C) 1.016 (32 - 46°C) 1.018 (32 - 46°C) 1.012 (20 - 32°C) 1.013 (32 - 46°C)	1.013 (32 – 46°C)	1.017 (32 – 46°C)
2.5	1.011 (20 – 32°C)	1.005 (20 – 32°C)	1.008 (20 – 32°C)	1.027 (20 – 32°C)
	1.008 (32 – 46°C)	1.008 (32 – 46°C)	1.013 (32 – 46°C)	1.016 (32 – 46°C)
4	1.003 (20 – 32°C)	1.011 (20 – 32°C)	1.008 (20 – 32°C)	1.026 (20 – 32°C)
	1.007 (32 – 46°C)	1.016 (32 – 46°C)	1.019 (32 – 46°C)	1.016 (32 – 46°C)
4.2	1.008 (20 – 32°C)	1.015 (20 – 32°C)	1.010 (20 – 32°C)	1.025 (20 – 32°C)
	1.013 (32 – 46°C)	1.018 (32 – 46°C)	1.019 (32 – 46°C)	1.015 (32 – 46°C)
Average	1.011 (20 – 32°C)	1.012 (20 – 32°C)	$1.009(20 - 32^{\circ}C)$	$1.028(20-32^{\circ}C)$
	1.008 (32 – 46°C)	1.013 (32 – 46°C)	1.015 (32 – 46°C)	1.016 (32 – 46°C)

Table 13 Temperature impact coefficients

* where,

T: target temperature

C: concentration (or volume) at the target temperature

t: reference temperature

 C_t : concentration (or volume) at the reference temperature (equation according to Metcalf & Eddy, 2003; the exponential equation indicates in practice the same percentage change per degree centigrade as the Arrhenius type of equation)

4.1.4 Discussion

From the LFG generation and utilisation point of view, the mesophilic condition is optimal for increasing methane utilisation efficiency, which is in accordance with the results of Blakey et al.

(1997) and Mata-Alvares and Martina-Verdure (1986). For small and medium-size landfills, attempts to maintain optimal temperatures for landfill operations are required. A bottom water insulating layer or recirculated leachate heating could be considered (Rees et al., 1980; Warith et al., 2005). However, if LFG collection and energy recovery were not required, a landfill temperature increase offers no significant environmental benefit when considering a long aftercare period.

When considering the post-closure management needs, under psychrophilic conditions, the residual COD concentrations with an L/S ratio of 4.3 (which corresponds to 200 years of landfilling in field conditions, as mentioned in Table 8) were close to the direct discharge limit, which is 200 mg/L. Under thermophilic and mesophilic conditions, the residual COD concentration is still clearly higher than the limit. In the case of nitrogen, the same trend can be seen, but the limit value is still far below the level achieved in this study, indicating the dominating status of nitrogen for the length of the aftercare period. Specific in situ leachate management and pretreatment processes (e.g. a nitrification-denitrification plus post DN system) are compulsory for countries with leachate indirect discharge limits.

It verifies that a higher temperature leads to greater and faster LFG emission. The LFG indicates most clearly the biological activity and is thus more sensitive to temperature change than other parameters (Cl, NH₄-N, COD), which have a stronger relation to physico-chemical phenomena such as washing.

This study indicates that the aftercare period may exceed 200 years before the leachate meets the mentioned limit values. This period can be shortened to some extent with a suitable leachate management and treatment system, including water recirculation, to achieve the target L/S ratio faster. To optimize aftercare from the cost point of view, different landfill management scenarios and leachate treatment alternatives have to be evaluated in relation to the circumstances in question, which is an extensive topic. These issues will be targeted in Section 4.4. As a whole the results give new information to evaluate applicable long-term strategies in various environmental conditions and for modelling. The long duration of the required aftercare period in anaerobic conditions indicates that other alternatives, like changing conditions at a suitable L/S ratio to a faster aerobic degradation, may also be useful, as shown by Heyer (2003).

4.2 Leachate biological treatment: nitrogen removal (III, IV)

The biological treatment of municipal landfill leachate was studied at 25 °C and with an average hydraulic retention time of 12 days. Without external carbon addition, the biodegradability of the raw leachate fluctuated and the BOD₇/N ratio was in the range of 0.1 - 0.5, which was comparatively low even for the aged landfills. In order to comprehensively evaluate the feasibility of decreasing the TN of landfill leachate with an on-site leachate pretreatment process, by applying low external carbon use and low DO, external carbon was added to escalate the OLR from the level of $0.01 \text{ g BOD}_{7/\text{g}}$ VSS/day to the level of $0.06 - 0.08 \text{ g BOD}_{7/\text{g}}$ VSS/day, with the NLR of 0.03 - 0.08 g N/g VSS/day. The operational schemes and the optimum BOD₇/N for denitrification were tested and compared as well.

4.2.1 Nitrification

In the nitrification phase, excessive carbon may cause an overgrowth of heterotrophic microorganisms that then interfere with the growth of autotrophic micro-organisms (Tijhuis et al., 1994), that is, ammonium-oxidizing bacteria, which similarly agglomerate on the sludge floc surfaces. In this study, the high influent NH₄-N concentration and progressive increase of external carbon level averted the occurrence of this inhibition.

A 99 – 100% average daily NH₄-N removal efficiency was achieved. During the aerobic phase, the maximum DO achieved was 2.0 mg/L and on average around 0.4 - 1.0 mg/L; the oxygen uptake rate (OUR) was measured at 4 - 13 mg O₂/L/hour and varied with different operational schemes. The applied C/N ratio (external carbon addition) was kept at the reasonably low level of 0.4 - 1.6. The nitrification rate was 2 - 4 mg/g VSS/hour. The results of Pelkonen et al., (1999) are close to the range found here, which was tested with a higher applied C/N ratio and DO level but at a lower temperature. Moreover, it was found that even with the DO lower than 0.4 mg/L, more than 98% NH₄-N removal efficiency could still be achieved stably. VFAs in the influent leachate can interfere with the nitrification (Eilersen et al, 1994). According to the DO and pH results, VFA probably concretized a big portion of BOD₇ represented in Table 3. Due to the limited amount of readily degradable organic content in the leachate, this may not have occurred in this study, which is supported by the near complete ammonium removal results. These results indicated the potential to reduce the aeration duration or to apply a low oxygen

supply as aeration. When the average DO was lower than 0.8 mg/L, the NO₂-N/NO_X-N ratio was around 0.7, which shows that with low DO and at a low external carbon addition level, it was possible to maintain an inhibition of nitrite-oxidising bacteria, leading to stable nitrite accumulation. More details about the DO control and aeration performance with different operational schemes will be discussed further in Section 4.2.4.

4.2.2 Denitrification

The highest average TN removal efficiency was achieved with an external carbon addition level of 1100 - 1800 mg COD/L, which was on average 55%.

Because of the variation in the leachate quality, the higher external carbon addition level does not necessarily represent a higher applied C/N ratio. To minimize this interference, the overall C/N ratio was intended to be used as a criterion of analysis, instead of calculating it based on explicit stages (external carbon addition levels). The optimal stable TN removal efficiency achieved was over 62% with the C/N ratio above 1.7. This TN removal efficiency is quite acceptable with such low C/N ratios, compared with the results shown for different (not only leachate) treatment processes (e.g. Ruiz et al. 2001; Obaja et al. 2005; etc.).

Even with a high C/N ratio, which was sometimes caused by the increase of slowly degradable organics or the decrease of NLR, lower OLR (meaning the lower availability of readily degradable organic matter (BOD₇)) still inhibited denitrification markedly. For example, as the C/N ratio increased from 1.4 to 1.6, a 1.5% decrease in the BOD loading rate induced a 4.6% consequential decrease in TN removal efficiency. This finding confirms that the availability of readily degradable organics determined the high point of biological treatability or biodegradability of the leachate (e.g. Peng et al., 2004).

A denitrification cyclic test was performed when the external carbon addition level reached 280 mg COD/L. The influent biodegradable COD/N ratio was 1.1, and the BOD/N ratio was half of that. This was designed intentionally with carbon limitation, in order to optimize the process so that costs are minimized, which will be discussed more in Section 4.3. With such low biodegradability of leachate, the average TN and COD removal efficiencies were 52% and 58%, respectively. With the intensive monitoring of several anoxic phases, the denitrification rate was determined to be 1.2 - 5.3 mg NO₃-N/(g VSS hour). In this case, it was detected that a BOD/N ratio greater than 0.9 is already sufficient to achieve a TN removal efficiency of over 60%.

Nevertheless, for stability of the treatment, it is still recommended that this ratio is higher than 1.2 (corresponding C/N level 1.7 - 1.8).

4.2.3 Partial nitrification and nitrite denitrification test period

Aiming at the characteristics of the landfill leachate, like high NH₄-N concentration and a low biodegradable COD/N ratio, a 120-day partial nitrification and nitrite denitrification test period (DO: < 1 mg/L and on average 0.4 - 0.6 mg/L, temperature: $26\pm1^{\circ}$ C and pH: 7.8 - 8.0) was performed.

Accumulation of nitrite is usually ascribed to the difference in the affectability of DO between ammonium oxidizing bacteria and nitrite oxidizing bacteria. In this condition, no evident inhibition to ammonium oxidization was observed and 71 - 89% of NH₄-N was converted to NO_2 . It can be deduced that at DO 1.2 mg/L or higher, complete nitrification to NO_3^- was achieved. When the DO was lower than 0.97 mg/L, the NO_2^-/NO_x^- ratio was greater than 0.5. These DO levels are lower than most of those reported by previous researchers (reviewed by Peng and Zhu, 2006). The low DO level showed no adverse effects on the ammonia oxidization rate or activated sludge quality. Briefly, controlling the aerobic DO level to below 1.0 mg/L promoted improved denitrification in the anoxic phase through nitrite accumulation. This indicates denitrification via nitrite at a lower DO. Accordingly, the specific nitrogen removal rate was highest at a NO₂ /NO_x ratio above 0.75, namely 23 mg N/(g VSS * d). The nitrite denitrification rate was high compared with the denitrification rate via nitrate, which is close to the results shown by van Kempen et al. (2001) and Chung and Bea (2002). With the same C/N ratio, the TN removal increased to a maximum 13% unit, corresponding to the 28% relative change during this period, which is in the range possible for partial nitrification and denitrification via nitrite.

Organic matter removal and carbon consumption during partial nitrification and nitrite denitrification test period

The organic matter removal has varied mostly with the organic loading and the lowest COD removal happened during the highest COD loads. With the C/N ratio above 1.7, the optimal COD removal efficiency achieved was 74%. When the DO level was under control, a decrease of 5 - 8% units in COD removal can be estimated at higher NO₂⁻/NO_x⁻ ratios.

Meanwhile the efficiency of the use of different carbon (COD) fractions can be seen clearly to be more efficient with an increasing NO₂ fraction of NO_x during the nitrite accumulation period and the preceding period, changing the conditions to being nitrite dominant. When the NO₂-fraction was over 0.75, the COD_{dosed}/N_{removed} ratio was 2.5, which is close to the value found in a Sharon process by Fux et al. (2003), namely 2.4 g COD _{added}/g NO₂-N _{removed} and slightly lower than that shown by Ruiz et al. (2006) in nitrite accumulation experiments. In this study the process conditions have been alternated oxic/anoxic, so some carbon loss could be expected. However, the ratio found here shows efficient carbon use, comparable to nitrite denitrification in permanent anoxic conditions. Possible additional factors aiding the low carbon need can be some uptake of carbon as cell internal storage compounds and the use of biodegradable COD in the leachate. In alternating carbon feed and anoxic/aerobic conditions with low DO, the readily degradable carbon is efficiently taken up as storage polymers and can be used for denitrification (Third et al., 2003).

As a whole, the biodegradable COD of raw leachate is 15 - 30% of dosed COD per one unit nitrogen removed, and it shows some increasing trend, together with the increasing nitrite fraction. Based on the anoxic biodegradation kinetics with leachate (Plosz et al., 2009) and substrate characterisation, it seems probable that part of the slowly degradable COD has been used for denitrification – before the next 4-hour period with new substrate feed. However, while part of this COD fraction has been consumed aerobically during the aerated period, this may have been minimised, especially during periods with a low DO set-point.

With a lower nitrite fraction of NO_x (below 0.3) the carbon need was clearly higher, approximately double compared to the range above 0.75. $COD_{dosed}/N_{removed}$ was close to 5.5 – 6, which is in the conventional range, as also found by Heander (2007) and slightly higher compared to the values found by Chang (1993, 1998).

As a result TN removal close to 60% was achieved at a low BOD/N ratio of 0.7 - 0.75, which made it possible to apply a lower external carbon addition to save costs and decrease residual COD concentrations.

4.2.4 **Operational strategies**

Two operational strategies and operational schemes, schemes PA and PB as illustrated in Figure 8, were run for 290 days and 308 days respectively. The main difference between schemes PA

and PB was the intervals between aeration; moreover, the PA scheme ran with a longer total anoxic period (2.5 hours/4-hour period) than PB (2.0 hours/4-hour period). As the biodegradable organics to nitrogen ratio (BOD₇/N ratio here) was around 1.2, removal efficiencies of 59% for TN and 55% for COD were achieved during scheme PA; in scheme PB, removal efficiencies were 61% for TN and 59% for COD. In scheme PB the TN removal efficiency was slightly better than during PA. Both of them are quite acceptable with the shown BOD₇/N ratio, which was kept low intentionally in order to have carbon limitation. COD removal was more efficient in scheme PB.

In this test, for both schemes PA and PB, the average DO was successfully modulated in the range of 0.5 - 1.0 mg/L. It was observed that at DO of 0.5 - 0.7 mg/L, a more effective inhibition of nitrite-oxidizing bacteria and nitrite accumulation happened.

Since DO control limitation was utilized in this study, the real time of aeration might be different from the design. A longer total non-aerated period occurred in PB in each cycle. At the same air supply level, a higher average nitrite concentration after aeration occurred in PB, which indicates that the longer total non-aerated period is advantageous to the inhibition of nitrite-oxidizing bacteria. In the PB scheme, the aeration period was shortened to 0.5 hour, which means that each non-aerated period was also short (maximum 1 hour).

Baumann et al. (1997) and Mota et al. (2005) studied different lengths of non-aerated periods and concluded that a reactor with more than 3-hour non-aeration period showed considerable nitrite reduction during the non-aeration periods and also reduced the amount of nitrite oxidizing bacteria. In this study the NH₄-N concentration in the influent was clearly higher and the DO concentration was considerable lower than in the study of Mota et al. (2005) – factors which favour the build-up of nitrite. Thus the control of DO could improve the selection of ammonium-oxidizing bacteria over nitrite-oxidizing bacteria in intermittently aerated schemes. The short aeration period (0.5 hour) didn't affect the virtual presence of the non-aerated period and showed even better performance at the inhibition of nitrite-oxidizing bacteria. In addition, it has been shown that high ammonium concentration is helpful in selecting ammonium-oxidizing bacteria over nitrite-oxidizing bacteria (Tappe et al., 1999). The influent feeding mode was significant in aiding ammonium accumulation during the non-aerated period, as the influent feed was at the beginning of the non-aerated period.

Under a low air supply condition, e.g. with the air supply level between 12 and 23 g O_2/g biodegradable COD and nitrogen (the indicator of air supply level), load PB showed a

promising adaptability with the accumulation of nitrite with up to 60 - 70% of nitrogen oxide and around 60% TN removal achieved stably. All nitrite ions were oxidized to nitrate ions when the air supply level was increased to 42 in PB and 54 g O₂/g biodegradable COD and nitrogen load in PA. With the same removal efficiency level, the supplied amount of air in PB was lower and showed better oxygen utilization efficiency; whereas with a higher air supply level (e.g. air supply level >30 g O₂/g biodegradable COD and nitrogen load), PA showed greater potential to prevent the nitrite oxidizing.

When the NO₂ fraction increased, the carbon needed for denitrification clearly decreased in both schemes and the biggest change happened when nitrite had started to accumulate (NO₂ fraction 0 - 0.3). As discussed in Section 4.2.3, the level of the COD_{dosed}/N_{removed} ratio in this study was rather promising. Similarly, in nitrite dominant conditions, the COD_{biodegradable}/N_{removed} ratio measured was lower than 3 in both PA and PB, which is close to values found in permanent anoxic conditions aimed at nitrite denitrification in spite of alternating anoxic/aerobic conditions.

The special operation strategies, like step-feeding and intermittent aeration, were effective at increasing the availability of readily biodegradable organics for exogenous denitrification during anoxic phases. Scheme PB showed better performance at effectively utilizing the aeration and preventing the nitrite from oxidizing. However, when considering the complexity of the real high-strength influent leachate features, the DO controlling strategy and measures are more demanding. It was found that the denitrification performance was improved as the aeration DO was kept at a low level: 0.4 - 0.6 mg/L. In the low DO condition, the extended anoxic phase and simultaneous nitrification and denitrification alleviated the adverse impacts of the inadequate anoxic time in both schemes.

4.2.5 Discussion

In general, the purpose of leachate biological treatment study is to verify the feasibility of improving leachate management in the aftercare period. For the decisive factor nitrogen (TN), the optimum removal targets with the best cost efficiency were determined via a series of experiments with different operational conditions (e.g. DO, the C/N ratio, the operational scheme). The results found were successful in complementing nitrogen removal with denitrification via nitrite and in achieving around 60% TN removal, a level which is reasonable

for a pretreatment option with strong carbon limitation.

With DO and scheme control, the carbon needed for denitrification clearly decreased, and the biggest change happened when nitrite had started to accumulate (NO₂ fraction 0 - 0.3). The level of the COD_{dosed}/N_{removed} ratio and the COD_{biodegradable}/N_{removed} ratio, measured in nitrite dominant conditions, were encouraging and were even lower than found by Pynaert et al. (2002), in a biofilm process with DO limitation, and lower than in the study of Monclus et al. (2009), with municipal landfill leachate in a SBR process. When denitrification takes place via nitrite it reflects a lower need for carbon (Zeng et al., 2003; Third et al., 2003) and makes the low cost considerations feasible. The experimental results are used for the cost estimation, which is discussed mainly in Section 4.3. The outcomes support the research purpose well and are crucial for the work on landfill leachate scenarios.

4.3 Cost estimation of leachate treatment (III, IV)

With a scale-up case study of the cost estimation based on the test landfill condition, the economic analysis of the on-site leachate pretreatment process was integrated as an important part of the study to define the applicability of the technology. The process developed in this study works effectively under low C/N and low DO conditions, which means that the operational cost for chemicals could be substantially reduced.

Currently, the leachate management costs are mainly due to the leachate treatment fee of the WWTP. After the on-site pretreatment process is adopted, the overall leachate management costs can be divided into two major parts: the leachate pretreatment costs (investment costs and general pretreatment operation/maintenance costs – mainly involving personnel, analytics and chemical costs) and the leachate treatment fee of the WWTP.

In the cost estimation, only the main cost items are included in order to compare the alternative costs rather than calculate the exact costs. In this estimation the measured performance values have been adapted to the full-scale conditions. The basic unit cost of pretreatment investment and general operational/maintenance was estimated to be $0.54 - 1.40 (0 - 90\% \text{ TN removal}) \text{ } \text{€/m}^3$, where the investment costs are variable with the treatment process capacity and reactor volume, and the operation costs (especially the chemical costs) are variable depending on the need of carbon addition and the TN removal target. The actual WWTP treatment fee was estimated to be $4.29 - 1.14 (0 - 90\% \text{ TN removal}) \text{€/m}^3$.

With 60% TN removal the cost saving potential is 28% and with 90% TN removal it is 38%, which indicates that the higher the TN removal in pretreatment, the higher the cost saving potential. When over 60% TN removal is targeted, the difference between the two options is rather small; the separate post DN (to increase TN removal from 60 to 90%) seems to be slightly less costly due to a lesser reactor volume and carbon need. If the whole process is working within the conventional range (e.g. Heander, 2007; Chang, 1993: $COD_{dosed}/N_{removed}$ – ratio 5.5 – 6 for 90% TN removal), the best cost saving potential is around 30%, achieved with 75 – 85% TN removal.

In a conventional two-stage system the disadvantages are that the influent biodegradable matter cannot be utilized in denitrification, and the decreasing pH in the nitrification unit must be compensated by a considerable amount of base addition. Both drawbacks increase chemical costs, which may be avoided using the option shown here. Moreover, in recent studies a reduced chemical (carbon) need has been found with denitrification via a nitrite pathway in two-stage systems (Canziani et al., 2006; Bai et al., 2009).

Referring to the discussion about landfill leachate management strategies in Section 3.6, the advanced post-treatment, like GAC filtration, was considered for the targets of direct discharge. The costs have been estimated as $4.2 \text{ } \text{e/m}^3$ for medium-sized landfills and $3.5 \text{ } \text{e/m}^3$ for big landfills, based partially on data provided by Theilen (1995) and Ivashechkin (2006).

The dominating role of the nitrogen load in wastewater treatment costs is a common feature in Nordic countries and elsewhere where the wastewater temperature is low during winter, resulting in higher investment costs. Therefore, the cost estimation in this study has wide reference value to other cases.

4.4 Landfill leachate management: nitrogen control and treatment (V)

4.4.1 Landfill emission potentials: NH₄-N, COD, chloride and LFG emission

Based on the results and discussion of leachate emissions in Section 4.1, the forecast models for the major emissions were established. The coefficients of the emission potential forecast models

are presented in Table 14 and the forecasting curves at the main reference temperatures are illustrated in Figure 11.

$y = y_t \theta^{(T-t)}$						
L/S ratio	Θ		Per cent error			
	Temperature	e ranges				
	20 – 32 °C	32 – 46 °C	20 – 32 °C	32- 46 °C		
NH ₄ -N	•	•	-			
0.6	1.017	1.014	0.3%	-0.4%		
1	1.014	1.012	0.5%	0.6%		
1.5	1.015	1.012	0.2%	-1.1%		
2	1.009	1.010	0.9%	1.3%		
2.5	1.005	1.008	-0.7%	-1.1%		
4	1.011	1.016	0.2%	-1.0%		
COD						
0.6	1.008	1.013	0.2%	0.9%		
1	1.011	1.013	-0.1%	0.6%		
1.5	1.007	1.013	0.2%	0.8%		
2	1.009	1.013	0.2%	-0.4%		
2.5	1.008	1.013	0.1%	0.2%		
4	1.008	1.019	-0.5%	-0.5%		
Chloride		·		-		
0.6	1.016	1.006	-0.2%	0.0%		
1	1.014	1.006	0.0%	0.5%		
1.5	1.013	1.006	0.3%	-0,3%		
2	1.009	1.009	-1.0%	0.7%		
2.5	1.011	1.008	0.5%	1.2%		
4	1.003	1.007	-0.6%	0.7%		
$y = ae^{bl}$			•	-		
Temperatures	А	b	R-squared value	P-value		
NH ₄ -N	•	•	• •	•		
20	1534.28	-0.5264	0.99	0.02		
32	1874.11	-0.5572	0.99	0.04		
46	2022.33	-0.5062	0.99	0.05		
COD	•	•	-			
20	3420.78	-0.7170	0.99	0.01		
32	3794.03	-0.7087	0.98	0.05		
46	4345.57	-0.6417	0.99	0.05		
Chloride			•	•		
20	2525.54	-0.5744	0.98	0.05		
32	2871.63	-0.6000	0.98	0.04		
46	2678.90	-0.5432	0.99	0.03		

Table 14 Coefficients of the leachate emission forecast models



Figure 11 Modelled leachate concentrations and gas production

The NH₄-N concentration in the leachate reaches the direct discharge limit value at an L/S ratio of 5.9 under psychrophilic and mesophilic conditions, and at an L/S ratio of 6.7 under thermophilic conditions. Meanwhile, the total amount of NH₄-N discharged in psychrophilic conditions is 13% and 28% less than the amount discharged in mesophilic and thermophilic conditions respectively. Leachate COD reaches the direct discharge limit value at an L/S ratio of 4.0 in psychrophilic conditions. In mesophilic conditions, the COD emission concentration meets the direct discharge limit value at an L/S ratio of 4.2; whereas in thermophilic conditions the required L/S ratio is 4.8. When meeting the direct discharge limit value, the total amount of COD discharged in psychrophilic conditions is 12% less than that in thermophilic conditions. For chloride emission, in psychrophilic and mesophilic conditions, the L/S ratio level required to meet the direct discharge limit value is

almost the same as well; that is, 5.6. In thermophilic conditions, the required L/S ratio to reach the direct discharge limit value is approximately 6.1. The total amount of chloride discharged is lowest in psychrophilic conditions and highest in thermophilic conditions (+13%).

It indicates that, compared to lower temperatures, the mesophilic conditions will not obviously increase the required L/S ratio needed to meet the limit value for the NH_4 -N emission, but will still increase the amount of NH_4 -N, which then must be taken care of.

The most active LFG generation and methanization kinetics occurred with an L/S ratio below 1, making it more difficult to forecast. The cumulative LFG generation was highest in the experimental range in thermophilic conditions, slightly lower in mesophilic conditions and clearly lowest in psychrophilic conditions – for which the results are in accordance with the leachate emission data. As LFG utilization is the target, increasing the landfill temperature is helpful in impelling waste degradation and methane generation and improving the efficiency and service life of the LFG utilization facilities. The forecasting model results also show that a lower L/S ratio rate can result in a lower target L/S ratio level, but will prolong the time needed to achieve the target L/S ratio level. At 20 °C, the required aftercare time is, in principle, prolonged by 10%; and, at 32 °C, the aftercare time, in principle, is doubled. It is clear that from the landfill aftercare point of view, a high L/S ratio rate is favourable.

It is expected that with the model developed here clues about the importance of different target parameters can be found. The importance may alter as the ranks of different discharge limits vary in different countries. According to the literature on the limit values (Table 12), the results shown can be applicable for many countries where similar discharge limit values are used. Varying recirculation strategies, such as fresh water and treated effluent circulation both in bioreactor and conventional landfills, can affect the behaviour of decisive factors (nitrogen, organic matters and inorganic matters). Based on the forecasting model results, the weighting coefficients of different factors for LE, referring to the equation in Section 3.6.2) are summarized in Table 15 and the LE trends at different temperatures are shown in Figure 12. There is a clear gap between the thermophilic conditions and the lower temperature conditions. When the temperature is lower than $32 \,^{\circ}$ C, the LE value decreases to 1 (direct discharge level), with an L/S ratio of 5.9 - 6.0. When the temperature is increased to a thermophilic level, an LE of 1 is achieved, with an L/S ratio of 6.7.

	per es,	L/S ratio	NH ₄ -N, mg/L			COD, mg/L			Cl ⁻ , mg/L		
		rate,	Direct	L/S	α	Direct	L/S	А	Direct	L/S	α
	em	/reference	discharge	ratio @		discharge	ratio @		discharge	ratio @	
	aı	year	limit	standard		limit	standard		limit	standard	
	20	1.175	70	5.9	0.38	200	4.0	0.26	100	5.6	0.36
20	20	0.860		5.6	0.41		3.4	0.25		4.5	0.34
	32	1.175		5.9	0.38		4.2	0.26		5.6	0.36
	32	0.411		6.0	0.41		3.4	0.23		5.1	0.36

Table 15 Weighting coefficients of leachate emission integrator



Figure 12 LE integrators at different temperatures

4.4.2 Post-closure management: a leachate management scenario and strategic model

Referring to the description in Section 3.6.3, six scenarios were designed and the major indictors are the size of landfill, represented by temperature difference, and the annual water infiltration, represented by annual L/S ratio difference. In order to make the results of the modelling work more widely applicable, the estimation of the landfill size and field conditions in this part (referring to Table 11) was different from the estimation in the previous part (referring to Table 8). This means that the simulated real landfill years resulted should not be compared. The LE integrator forecasts for different scenarios, illustrated in Figure 13, shows the different time frames needed to achieve the target L/S ratio.

For big landfills with the basic Scenario A, the aftercare period may be as long as 440 years. With three-fold infiltration (Scenario B), this period would be approximately 145 years. When six-fold infiltration is used (Scenario C), the aftercare period for leachate could be shortened to approximately 75 years. For medium-sized landfills, the estimated aftercare period for scenarios E, F, and G are approximately 145, 50 and 25 years respectively. Scenarios C and G provide the best starting point for highlighting the importance of recirculation. Moreover, the length of these periods refers to conventional landfills and, if operated as bioreactors from the beginning, shorter periods would result.



Figure 13 LE trends in different scenarios

Due to recirculation, the biodegradation of organic material in landfill is enhanced, as has been verified in simulator experiments, which is in accordance with the results of Lema et al. (1988) and Jiang et al. (2007). However, recirculation is not effective in reducing the NH₄-N concentration; on the contrary, it increases the NH₄-N concentration to some extent (Benson et al., 2007). Hence, it is favourable to nitrify the NH₄-N and to reduce the total nitrogen concentration through an on-site pretreatment process. Nitrification is more important, since denitrification may happen in the anaerobic conditions of inner landfills together with organic waste degradation. Denitrification is also helpful in accelerating the L/S ratio rate compared with the recirculation of untreated leachate.

According to these understandings, a cost estimation is made for different scenarios based on the near-real situations of Finnish landfills and applied only to the aftercare period. Chlorides are not considered as a decisive factor because of their negligible role in the salt load of the municipal wastewater system. In the case of on-site treatment, the recipient can be the same as with the WWTP, keeping the salt load comparable for different leachate management alternatives. The results show that a single biological treatment, with a low addition of carbon, decreases operational costs. The total costs, including capital, operational and maintenance costs, as well as the average costs per m³ of leachate and per ton of landfilled waste, are shown in Figure 14.



Figure 14 Estimated total and average costs in different scenarios and leachate management strategies (The vertical bars indicate the differences of the total costs, with \pm 25% unit cost

changes for on-site advanced treatment and the WWTP treatment fee considered in the cost estimation.)

In this estimation, post DN with advanced treatment was considered when the effluent's COD does not meet the limit for on-site leachate management alternatives. GAC filtration is a good option as an advanced treatment method in the sense that the cost is mostly related to carbon material and is not as much connected to plant size, whereas other methods have a much higher percentage of capital costs. This makes the estimation of this treatment alternative more concrete and more actual.

In Finland, if a municipal treatment plant is available and has an adequate capacity, it is customary to treat the leachate at the existing public facility (Off-site I in Figure 14). With the current leachate management approach in Helsinki (municipal WWTP), the high NH₄-N concentration corresponds to approximately 65% of the unit costs. It can be assumed that in many existing landfills, an indirect discharge may be prioritized; however, the costs of this indirect discharge strategy without on-site pretreatment (Off-site I management alternative) are the highest. Moreover, the total cost of Off-site I was the most sensitive to the fluctuation of estimated unit costs (indicated by the vertical bars in Figure 14), which means uncertainty for the long-term management strategies.

Depending on the leachate characteristics and the local indirect discharge regulations, an on-site leachate pretreatment process should be considered for the indirect discharge. On-site biological pretreatment with post DN (Off-site III management alternative) shows the lowest costs in all scenarios, which reflects the importance of nitrogen removal. When minimizing the length of the aftercare period, on-site treatment (On-site management alternative) with activated carbon filtration shows clear advantages, but it is to some extent more expensive than the alternatives.

When comparing the different alternatives in the same scenario related to possible changes in the estimated unit costs, the costs of Off-site III are still the lowest (Figure 14). Scenario C is similar to Scenario G, except that the difference in the on-site treatment alternative is a little smaller. This order prevails, excluding the base scenarios (scenarios A and E), in which the order of the different treatment alternatives may change with a small fluctuation in the unit costs. If temperatures differ from the estimated temperature, for example being 5 - 10 °C higher, then the impact on the cost comparison is no bigger than the estimated changes in the unit costs. It is worth emphasizing that local circumstances have a strong impact on costs and should always be considered.

In biological treatment, investment costs form the main part of the unit cost and it is estimated that they will increase 1.7-fold when the flow rate decreases one decade, which is considerable. There is a rather small difference in unit costs between a big landfill versus a medium-sized landfill, as in scenarios C and G. In a medium-sized landfill, the average costs even seem to be slightly lower than in a big landfill. The factors affecting this have to do with the fact that the difference in the flow rate is not very big (twofold) and that the initial NH₄-N concentration in a medium-sized landfill is estimated to be 14% lower than in a big landfill when the aftercare period starts. The latter brings some advantages by decreasing the total amount of leachate in favour of medium-sized landfills. Overall, it is possible to combine the environmental benefits (the shortest aftercare) with the economic benefits.

Berge et al. (2009) estimated that leachate treatment costs will decrease considerably during the aftercare period when leachate is re-circulated, assuming that only the leachate fraction not recirculated requires treatment. This is not supported by the simulator results in this study, where, conversely, the treatment steps shown in Figure 9 are needed when anaerobic bioreactors are considered. They used a 30-year period in their study, and so a comparison of the costs estimated here would only be partial.

The experiments by Fellner et al. (2009) showed that leaching related to soluble components like chloride can be different in full-scale situations as compared to simulator results. On the other hand, nitrogen leaching was comparable at full-scale and at a simulator scale. As nitrogen is the factor that determines the targeted L/S ratio, uncertainty regarding the estimated costs seem not to be serious, based on the simulator study results. It is also obvious that the structural properties of the leachate infiltration system have an impact on the full-scale leaching efficiency. Some uncertainty is to be expected, but the leached loads are the same for both onsite and off-site treatment alternatives. However, when considering interest rates for long-term cost estimation and comparison, divergent impacts on the scenarios are to be expected.

When using a high infiltration rate in medium-sized landfills, the effluent limit values can be reached in a reasonable time. For big landfills, this is not the case and other means, like the inclusion of an aerobic phase, are necessary. This will include additional investment and operational costs, but it does not change the total costs dramatically (Stegmann et al. 2006).

4.4.3 Discussion

With the studied emission limits, nitrogen (NH₄-N) is the key decisive factor with the longest aftercare time. It corresponds to the results in Section 4.1 and the findings of Andreas (2000) and Morris and Barlaz (2011), and with such a long time required in principle for the aftercare, a specific treatment and management process for nitrogen is obligatory. To avoid Cl⁻ becoming the limiting parameter, fresh water addition must be involved in leachate recirculation, because in biological leachate treatment Cl⁻ removal is negligible. The tendency of temperature impacts and L/S impacts showed by the forecasting model is also in accordance with the findings of the landfill simulators in Section 4.1.

The scenario results for leachate emissions reveal that the targeted L/S ratio required to end the landfill leachate aftercare is 5.7 - 6 and a 30 - 40-year aftercare period is not sufficient for all scenarios. The L/S ratios needed to achieve the limit values found here are to some extent higher, on average, than the values expressed by Heyer et al. (2005). They found that the highest L/S ratios needed for nitrogen are 4.4 on average (maximum 7.7), giving an over 200-year time period (450 years maximum). The difference in results between Heyer et al. (2005) and this study is caused by a lower organic fraction and some differences in infiltration and landfill size.

The highest water infiltration (Scenario C) in a big landfill resulted in the shortest aftercare period. However, it may be technically problematic to achieve a high infiltration rate, and specific measures should be taken, some of which may be more costly than in other cases. With lower waste depths (Scenario G), such infiltration rates can be achievable, although such full-scale realizations may still be scarce.

On-site biological pretreatment seems to be effective from both technical and economical points of view and is recommended. Especially when leachate recirculation has been adopted, a considerable increase in plant size and a shorter length for the aftercare period shows a clear advantage in unit costs.

The average unit cost (per m³ leachate) found here (Figure 14) are lower than those found by Heyer et al. (2005). The period for which they estimated the costs is obviously shorter, because the total costs found here are higher. The lower costs (per m³ leachate) here may be due to the selection of treatment techniques being directed at low-cost alternatives. Also, the average costs here include periods during which some unit process(es) may not be operating and, at the earlier stage, the costs may have been higher. Compared with the calculation for a conceptual sample

landfill by Stegmann et al. (2006), the leachate management costs are in the same range (in relation to the waste volume) when the lowest costs (Scenario G) are considered.

According to the estimated leachate management costs shown in Figure 14, it is possible to achieve a reasonable cost reduction of 40% in big landfills and 34% in medium-sized landfills by the improvement of leachate management strategies. This output is meaningful to answer the research questions raised in Chapter 1.

Validated by the intensive simulation data, the models developed in this study forecast the potentials of the leachate emissions very well. This outcome makes it possible to determine the length of the landfill aftercare period.

The forecasting results of different decisive factors give hints of their importance and also the weighting coefficients to achieve LEs. The result of LEs principally embodies the specific decisive factors (nitrogen, organic matters and inorganic matters). With this tool, it is possible to use a single integrated indicator to study the feasibility of leachate management scenarios.

The model developed here is targeted to be an effective prognosis tool for the indication of biodegradation performance (leachate emission) and for the development of the leachate management strategies system of real landfills. This methodology takes into account the whole lifespan of landfill leachate management rather than only roughly estimating it.

The approach is based on the LSR data and the site-specific conditions of full-scale landfills such as complex waste composition and local climate are not considered. So the model developed is facing limitations as to the efficiency (evenness) of infiltration and the heterogeneity of the waste. The uncertainty of the models needs to be discussed in the further study in view of the sensitivity and utility of model outcomes.

5 Conclusions

Leachate characteristics and emission potentials in the aftercare period

The results from the long-term anaerobic LSR showed that the impacts of temperature emerged clearly in all the parameters studied. At higher temperatures, waste degradation was more effective, which is favourable from the LFG management point of view (i.e. more LFG is generated). The mesophilic condition showed the optimal efficiency. However, from the leachate management point of view, a higher temperature also resulted in higher emission concentrations and landfill leachate emission potentials, which means a greater effort in leachate treatment is required. After an L/S of over 4, the waste degradation degrees of the mesophilic condition. Therefore, the increase in temperature can accelerate waste degradation and gas generation, but cannot decrease the length of the waste stabilisation since it could extend the waste degradation degree. A higher annual L/S ratio rate was verified to be an effective factor in considerably decreasing the stabilisation period.

The results of this study systematically describe the long-term landfill leachate and LFG emission performance, but the limit value is still far below the level achieved in the simulators with the achieved L/S ratio of 4.35. Even though the results cannot directly answer a question like the length of the landfill aftercare period, they give a solid foundation for the estimation of long-term landfill leachate emissions in different environmental conditions and forecast modelling.

Leachate treatment strategy to improve leachate management in the aftercare period

The results of biological leachate nitrogen removal studies showed the feasibility of on-site leachate biological pretreatment for treating a high NH₄-N concentration and low biodegradable COD/N ratio municipal landfill leachate. The denitrification performance was improved when the aeration DO was kept at a low level of 0.4 - 0.6 mg/L. Over 99% ammonium nitrification and at least 40% partial nitrification to nitrite could be achieved with a BOD₇/N of 1.1, and over 60% TN removal could be achieved with a reasonably low air supply level. In the low DO condition, the extended anoxic phase, and simultaneous nitrification and denitrification, alleviated the adverse impacts of the inadequate anoxic time and increased the probability of utilizing slowly degradable organics for denitrification.

The reaction rate of denitrification (both NO_2^- and NO_3^-) was 4.1 - 7.0 mg/g VSS/hour during scheme PA (long non-aerated/aerated sequences) and 3.2 - 6.0 mg/g VSS/hour during scheme PB (short sequences). The PB scheme performed better when saving air supply and preventing nitrite oxidation under low air supply conditions; it also showed, on average, 5% higher units of TN removal and better COD removal. Both tested schemes were successful in complementing nitrogen removal with denitrification via nitrite and in achieving around 60% TN removal, which is reasonable as a pretreatment option with strong carbon limitation.

The results are applicable to optimizing the low-cost operational strategies of an on-site pretreatment process. The experimental results make comprehensive cost estimation feasible and the outcomes support the evaluation of leachate management principles and strategies well, from the technical and economic points of view; something that is functional in detecting the essentials – the length and costs of leachate aftercare.

The methodology of landfill leachate management: forecast modelling and post-closure management

Based on the results above, the possibility and feasibility of optimizing landfill leachate management and treatment strategies have been clarified, from both a technical and economic point of view. As a guideline system to integrate the results and solve the questions, the forecast models for major emissions and their integrator (LE) were developed by using a regression analysis tool.

The results expressed the importance of different target parameters and gave clues to the target L/S ratio level of landfill needed in order to end leachate management. It indicated the dominating status of nitrogen in determination of the length of aftercare period, and thus nitrogen is the key indicator of long-term leachate performance. This verified the necessity of on-site leachate management and pretreatment processes, e.g. biological nitrification-denitrification, for the removal and control of the TN of leachate for different purposes, i.e. indirect discharge, leachate recirculation and/or on-site advanced leachate treatment. The targeted L/S ratio required to end the landfill leachate aftercare was revealed as 5.7 - 6, which was determined by studying waste characteristics (the organic fraction) and landfill conditions (infiltration and landfill size).

Most importantly, the tool makes the evaluation of the level of leachate emissions at a certain time possible, in order to directly give us an answer for the length of the aftercare period for a landfill. Six scenarios are designed, based on the general condition of existing sanitary landfills, to represent the medium-sized and big landfills, with or without recirculation. The forecast of six landfill-scenarios showed that in scenarios with the highest infiltration leachate, the aftercare period can be shortened substantially to 25 years and 75 years, in medium-sized and big landfills respectively. However, this is technically more challenging to do for big landfills. Moreover, the leachate management costs during the aftercare period can be reduced by 30 - 40% compared with conventional landfill leachate management.

Due to leachate characteristics and local indirect discharge regulations, the alternatives of onsite leachate pretreatment processes are deliberated and show economic advantages compared with the customary leachate management alternative (channelling to the WWTP). Because of the importance of nitrogen removal, on-site biological pretreatment with post DN showed the lowest costs in all scenarios. On-site treatment with activated carbon filtration is competitive within the shortened aftercare period as the alternative with the next lowest costs. Overall, the environmental benefits (the shortest aftercare period) and the economic benefits could be integrated based on the forecast modelling and cost analysis.

This methodology is expected to give landfill leachate management some outlook with the support of quantifiable data. Temperature and the L/S ratio are deliberated to represent the variety of landfill conditions and operation. The methodology is targeted to be widely applicable. It can estimate the length of the aftercare period feasibly and give the data support to solve the research questions raised in the beginning. The finding is significant for the decision makers of landfills in considering how to shorten the aftercare period and make cost plans.

References

Abufayed A. A., Schroeder E. D., 1986. Performance of SBR/denitrification with primary sludge carbon source. Journal Water Pollution Control Federation, 58: 387-397.

Andreas L., Bilitewski B., 1999. Effects of waste quality and landfill technology on the long-term behavior of municipal landfills. Waste Management and Research, 17: 413-423.

Andreas L., 2000. Langzeitemissionsverhalten von Deponien fuer Siedlungsabfälle in den neuen Bundesländern. Dissertation, Technische Universität Dresden.

Andreottola G., Foladori P., Ragazzi M., 2001. On-line control of a SBR system for nitrogen removal from industrial wastewater, Water Science and Technology, 34: 93-100.

Bae J. H., Kim S. K., Chang H. S., 1997. Treatment of landfill leachates: ammonia removal via nitrification and denitrification and further COD reduction via Fenton's treatment followed by activated sludge. Water Science and Technology, 36: 341-348.

Bai T., Lei H., Yu G., Yu Q., Li Z., Li H., 2009. High nitrite accumulation and strengthening denitrification for old-age landfill leachate treatment using an autocontrol two-stage hybrid process. Process Safety and Environmental Protection, 87(5): 307-314.

Barlaz M. A., Ham R. K., Schaefer D. M., 1992. Microbial chemical and methane production characteristics of anaerobically decomposed refuse with and without leachate recycling. Waste Management & Research, 10(3): 257-267.

Baumann B., Snozzi M., van der Meer J. R., Zehnder A. J. B., 1997. Development of stable denitrifying cultures during repeated aerobic-anaerobic treatment periods, Water Research, 31: 1947-1954.

Benson C. H., Barlaz M. A., Lane D. T., Rawe J. M., 2007. Practice review of five bioreactor/recirculation landfills. Waste Management, 27: 13-29.

Berge N. D., Reinhart D. R., Townsend T. G., 2005. The fate of nitrogen in bioreactor landfills. Critical Reviews in Environmental Science and Technology, 35: 365-399.

Berge N. D., Reinhart D. R., Dietz J., Townsend T., 2006. In situ ammonia removal in bioreactor landfill leachate. Waste Management, 26: 334-343.

Berge N. D., Reinhart D. R., Batarseh E. S., 2007. Strategy for complete nitrogen removal in bioreactor landfills. Journal of Environmental Engineering, 133(12): 1117-1125.

Berge N. D., Reinhart D. R., Batarseh E.S., 2009. An assessment of bioreactor landfill costs and benefits. Waste Management, 29: 1558-1567.

Bernet N., Dangcong P., Delgenes J., Moletta R., 2001. Nitrification at low oxygen concentration in biofilm reactor, Journal of Environmental Engineering, 127: 266-271.

Blakey N. C., Bradshaw K., Reynolds P., Knox K., 1997. Bioreactor landfill - a field trial of accelerated waste stabilization. Sixth International Landfill Symposium, S. Margherita di Pula, Cagliari, Italy, October 13-17, 375-386.

Boltze U., de Freitas M. H., 1997. Monitoring gas emissions from landfill sites. Waste Management and Research, 15: 463-476.

Boni M. R., Chiavola A., Sbaffoni S., 2006. Pretreated waste landfilling: relation between leachate characteristics and mechanical behaviour. Waste Management, 26: 1156-1165.

Buttol, P., Masoni, P., Bonoli, A., Goldoni, S., Belladonna, V., Cavazzuti, C., 2007. LCA of integrated MSW management systems: case study of the Bologna district. Waste Management, 27: 1059–1070.

Brock T. D., Madigan M. T., 1991. Biology of microorganisms, Sixth edition. Prentice-Hall International Editions, Inc.

Canziani R., Emondi V., Garavaglia M., Malpei F., Pasinetti E., Buttiglieri G., 2006. Effect of oxygen concentration on biological nitrification and microbial kinetics in a cross-flow membrane bioreactor (MBR) and moving-bed biofilm reactor (MBBR) treating old landfill leachate. Journal of Membrane Science, 286(1-2): 202-212.

Chang, L., 1993. Stickstoffelimination mit einstufigen Belebungsanlagen zur Behandlung von Sickerwasser aus Hausmülldeponien. In: Kayser R. (ed) Sickerwasser aus Hausmülldeponien – vermeiden- vermindern-behandeln. Veröffentlichungen des Zentrums fuer Abfallforschung der Technischen Universität Braunschweig, Heft 8, 135-162.

Chang, L., 1998 Auslegung von einstufigen Belebungsanlagen zur Stickstoffelimination bei Sickerwässern aus Siedlungsabfalldeponien. Dissertation. TU Braunschweig, Germany.

Christensen T. H., Cossu R., Stegmann R., 1992. Landfilling of waste: leachate, Elsevier Applied Science.

China Environmental Science Institute, 2007. National emission standards of municipal landfills, China, Draft version (in Chinese).

Chung J. and Bae W., 2002. Nitrite reduction by a mixed culture under conditions relevant to shortcut biological nitrogen removal. Biodegradation, 13(3): 163-170.

Egli K., Fanger U., Alvarez P. J., Siegrist H., van der Meer J. R., Zehnder A. J., 2001. Enrichment and characterization of an anammox bacterium from a rotating biological contactor treating ammonium-rich leachate. Archives of Microbiology, 175: 198-207.

Eilersen A. M., Henze M., Kloft L., 1994. Effect of volatile fatty acids and trimethylamine on nitrification in activated sludge. Water Research, 28(6): 1329–1336.

El-Fadel M., Findikakis A. N., Leckie J. O., 1997a. Environmental impacts of solid waste landfilling. Journal of Environmental Management, 50: 1-25.

El-Fadel M., Findikakis A. N., Leckie J. O., 1997b. Modeling leachate generation and transport in solid waste landfills. Environmental Technology, 18(7): 669-686.

El-Fadel M., Massoud M., 2001. Methane emissions from wastewater management. Environmental Pollution, 114(2):177-185.

El-Fadel M., Bou-Zeid E., Chahine W., Alayli B., 2002. Temporal variation of leachate quality from pre-sorted and baled municipal solid waste with high organic and moisture content. Waste Management, 22: 269-282.

Eden, R., 2000. Removal of ammonia from landfill leachate in Hong Kong. Organics Ltd.

European Commission, 1975. Council Directive of 15 July 1975 on waste (75/442/EEC). European Commission, Brussels, Belgium.

European Commission, 1991a. Council Directive of 18 March 1991 amending Directive 75/442/EEC on waste (91/156/EEC). European Commission, Brussels, Belgium.

European Commission, 1991b. Council Directive of 23 December 1991 standardizing and rationalizing reports on the implementation of certain Directives relating to the environment (91/692/EEC). European Commission, Brussels, Belgium.

European Commission, 1991c. Council Directive of 12 December 1991 on hazardous waste (91/689/EEC). European Commission, Brussels, Belgium.

European Commission, 1994. European Parliament and Council Directive of 20 December 1994 on packaging and packaging waste (94/62/EC). European Commission, Brussels, Belgium.

European Commission, 1996. Commission Decision of 24 May 1996 adapting Annexes IIA and IIB to Council Directive 75/442/EEC on waste (text with EEA relevance) (96/350/EC). European Commission, Brussels, Belgium.

European Commission, 1997. Council resolution of 24 February 1997 on a community strategy for waste management. European Commission, Brussels, Belgium.

European Commission, 1999. The European Council Directive on the landfill of waste (1999/31/EU). European Commission, Brussels, Belgium.

European Commission, 2000. Directive 2000/76/EC of the European Parliament and of the Council of 4 December 2000 on the incineration of waste. European Commission, Brussels, Belgium.

European Commission, 2004. Directive 2004/12/EC of the European Parliament and of the Council of 11 February 2004 amending Directive 94/62/EC on packaging and packaging waste. European Commission, Brussels, Belgium.

European Commission, Eurostat, 2011. Waste Statistics. WWW pages on Eurostat (http://epp.eurostat.ec.europa.eu/statistics_explained/index.php/Waste_statistics#Non-mineral_waste_generation).

European Standard, 1998. Water Quality - Determination of Biochemical Oxygen Demand after n Days (BOD_n) - Part 1: Dilution and seeding method with allylthiourea addition (ISO 5815:1989, Modified). EN 1899 – 1.

European Union, 1999. Council Directive 1999/31/EC on the landfill of waste. Official Journal of the European Communities.

Farquhar G. J., 1989. Leachate: production and characterization. Canadian Journal of Civil Engineering, 16: 317-325.

Fellner J., Döberl G., Allgaier G., Brunner P. H., 2009. Comparing field investigations with laboratory models to predict landfill leachate emissions. Waste Management, 29: 1844-1851.

Ferguson C. C., 1993. A hydraulic model for estimating specific surface area in landfill. Waste Management and Research, 11(3): 227-248.

Fux, C., Lange, K., Faessler, A., Huber, P., Grueniger, B. & Siegrist, H., 2003. Nitrogen removal from digester supernatant via nitrite – SBR or Sharon? Water Science and Technology 48 (8): 9-18.

Fux C., Siegrist H., 2004. Nitrogen removal from sludge digester liquids by nitrification / denitrification or partial nitritation / anammox: environmental and economical considerations. Water Science and Technology, 50(10): 19-26.

Fux C., Velten S., Carozzi V., Solley D. and Keller J., 2006. Efficient and stable nitrification and denitrification of ammonium-rich sludge dewatering liquor using a SBR with continuous loading. Water Research, 40(14): 2675 – 2775.

Galloway J. N., 1998. The global nitrogen cycle: changes and consequences. Environmental Pollution, 102: 15-24.

Garrido J., van Benthum W., van Loosdrecht M., Heijnen J., 1997: Influence of dissolved oxygen concentration on nitrite accumulation in a biofilm airlift suspension reactor, Biotechnology and Bioengineering, 53: 168-178.

Giannis A., Makripodis G., Simantiraki F., Somara M., Gidarakos E., 2008. Monitoring operational and leachate characteristics of an aerobic simulated landfill bioreactor. Waste Management, 28: 1346-1354.

Gioannis G. D., Muntoni A., Cappai G., Milia S., 2009. Landfill gas generation after mechanical biological treatment of municipal solid waste. Estimation of gas generation rate constants. Waste Management, 29: 1026-1034.

Grunditz C., Dalhammar G., 2001. Development of nitrification inhibition assays using pure cultures of Nitrosomonas and Nitrobacter. Water Research, 35 (2): 433 – 440.

Haarstad K., Mæhlum T., 1999. Important aspects of long-term production and treatment of municipal solid waste leachate. Waste Management and Research, 17: 470-477.

Hanson J., Yesiller N., Oettle N., 2010. Spatial and temporal temperature distributions in municipal solid waste landfills. Journal of Environmental Engineering, 136(8): 804-814.

Hartz K. E., Klink R. E., and Ham R. K., 1982. Temperature effects: methane generation from landfill samples. Journal of Environmental Engineering Division, Proceedings of the American Society of Civil Engineers, 108: 629-638.

Heander, E., 2007. The potential of nitrogen removal from leachate from Spillepeng with SBRtechnology and supplemental COD–reduction with chemical oxidation agents. MSc-thesis. Lund University, Sweden.
Hellinga C., Schellen A. A. J. C., Mulder J. W., van Loosdrecht M. C. M., Heijnen J. J., 1998. The SHARON process: an innovative method for nitrogen removal from ammonium-rich waste water. Water Science and Technology, 37:135-142.

Henze M., Harremoes P., la Cour Jansen J., Arvin E., 2002. Wastewater treatment. Biological and chemical processes. Springer Verlag Berlin.

Heyer K-U, 2003. Emissionreduzierung in der Deponienachsorge (In German). Hamburger Berichte 21, Abfallwirtschaft, Technische Universität Hamburg-Harburg.

Heyer K-U, Hupe K, Stegmann R., 2005. Landfill aftercare – scope for actions, durations, costs and quantitative criteria for the completion. In: Proceedings Sardinia 2005 Waste Management and Landfill Symposium, Sardinia pp. 11, Italy.

Hoilijoki T. H., Kettunen R. H., Rintala J. A., 2000. Nitrification of anaerobically pretreated municipal landfill leachate at low temperature. Water Research, 34:1435-1446.

Hossain M. S., Gabr M. A., Barlaz M., 2003. Relationship of Compressibility Parameters to Municipal Solid Waste Decomposition. Journal of Geotechnical and Geoenvironmental Engineering, 129(12): 1151-1158.

Ilies P., Mavinic D. S., 2001. The effect of decreased ambient temperature on the biological nitrification and denitrification of a high ammonia landfill leachate. Water Research, 35:2065-2072.

Isermann K., Isermann R., 1998. Food production and consumption in Germany: N flows and N emissions. Nutrient Cycling in Agroecosystems, 52: 298-301.

ISO International Standard, 2005. Water Quality – Determination of Ammonium Nitrogen – Method by Flow Analysis (CFA and FIA) and Spectrometric Detection, ISO 11732.

Ivashechkin P., 2006. Elimination of organic trace pollutants from municipal wastewater. Dissertation Technische Hochschule Aachen.

Jetten M., Wagner M., Fuerst J., 2001. Microbiology and application of the anaerobic ammonium oxidiation ('anammox') process. Current Opinion in Biotechnology, 12(3): 283-288

Jiang J. G., Yang G. D., Deng Z., Huang Y. F., Huang Z. L., Feng X. M., Zhou S. Y., Zhang C. P., 2007. Pilot-scale experiment on anaerobic bioreactor landfills in China. Waste Management, 27: 893-901.

Jokela J. P. Y., Kettunen R. H., Rintala J. A., 2002a. Methane and leachate pollutant emission potential from various fractions of municipal solid waste (MSW): Effects of source segregation and aerobic treatment. Waste Management and Research, 20: 434-444.

Jokela J. P. Y., Kettunen R. H., Sormunen K. M., Rintala J. A., 2002b. Biological nitrogen removal from municipal landfill leachate: low-cost nitrification in biofilters and laboratory scale in-situ denitrification. Water Research, 36: 4079-4087.

Jokela J. P. Y., Rintala J. A., 2003. Anaerobic solubilisation of nitrogen from municipal solid waste (MSW). Re/Views in Environmental Science & Bio/Technology, 2: 67-77.

Kabdaşli I., Şafak A., Tünay O., 2008. Bench-scale evaluation of treatment schemes incorporating struvite precipitation for young landfill leachate. Waste Management, 28: 2386-2392.

Kalyuzhnyi, S., Sklyar, Y., Fedorovich, Y., Kovalev, A., Nozhevnikova, A. and Klapwijk, A., 1999. The development of biotechnological methods for utilization and treatment of diluted manure streams. Water Science and Technology, 40(1): 223-229.

Kettunen R. K., 1997. Treatment of landfill leachates by low temperature anaerobic, sequential anaerobic–aerobic processes. Ph.D. dissertation, Tampere University of Technology, Finland.

Kim J. H., Chen M. X., Kishida N. H., Sudo R., 2004. Integrated real-time control strategy for nitrogen removal in swine wastewater treatment using sequencing batch reactors. Water Research, 38: 3340-3348.

Kjeldsen P., Barlaz M. A., Rooker A. P., Baun A., Ledin A., Christensen T. H., 2002. Present and long-term composition of MSW landfill leachate: a review. Critical Reviews in Environmental Science and Technology, 32(4): 297-336.

Komilis D. P., Ham R. K., Stegmann R., 1999a. The effect of municipal solid waste pretreatment on landfill behavior: a literature review. Waste Management and Research, 17: 10-19.

Komilis D. P., Ham R. K., Stegmann R., 1999b.The effect of landfill design and operation practices on waste degradation behavior: a review. Waste Management and Research, 17: 20-26.

Kouvo, P., 2005. Landfill Gas Utilization in District Heating System. ISWA World Congress 2005, 6–10 November, 2005, Buenos Aires, Argentina.

Kurniawan T. A., Lo W. H., Chan G.Y.S., 2006. Physico-chemical treatments for removal of recalcitrant contaminants from landfill leachate. Journal of Hazardous Materials, 129(1-3): 80-100.

Laner D., 2011. Understanding and evaluating long-term environmental risks from landfills. Ph.D. dissertation, Vienna University of Technology, Austria.

Lema J. M., Mendez R., Blazquez R., 1988. Characteristics of landfill leachates and alternatives for their treatment: A Review. Water, Air, and Soil Pollution, 40: 223-250.Mace S., Mata-Alvarez J. R., 2002. Utilization of SBR technology for wastewater treatment: an overview. Industrial and Engineering Chemistry Research, 41: 5539–5553.

Manfredi S., Niskanen A., Christensen T. H., 2009a. Environmental assessment of gas management options at the Old Ämmämsuo landfill (Finland) by means of LCA-modeling (EASEWASTE). Waste Management, 29: 1588-1594.

Manfredi S., Christensen T. H., 2009b. Environmental assessment of solid waste landfilling technologies by means of LCA-modeling. Waste Management, 29: 32-43.

Mata-Alvares J., Martina-Verdure A., 1986. Laboratory simulation of municipal solid waste fermentation with leachate recycle. Journal of Chemical Technology and Biotechnology, 36: 547-556.

McBean E. A., Syed-Ritchie S., Rovers F. A., 2007. Performance results from the Tucumán solid waste bioreactor. Waste Management, 27: 1783-1791.

Mehta R., Barlaz M. A., Yazdani R., Augenstein D., Bryars M., Sinderson L., 2002. Refuse decomposition in the presence and absence of leachate recirculation. Journal of Environmental Engineering, 128(3): 228-236.

Mertoglu B., Calli B., Inanc B., Ozturk I., 2006. Evaluation of in situ ammonia removal in an aerated landfill bioreactor. Process Biochemistry, 41: 2359-2366.

Metcalf & Eddy, 2003 Wastewater engineering, treatment and reuse. 4th edition. McGraw-Hill, New York.

Ministry of the Environment, 2002. Revised national waste plan for the period up to 2005. WWW pages on Waste Policies (<u>http://www.ymparisto.fi</u>).

Monclus H., Puig S., Coma M., Bosch A., Dolors Balaguer M., Colprim J., 2009. Nitrogen removal from landfill leachate using the SBR technology. Environmental Technology, 30(3): 283-290.

Morris J. W. F., Barlaz M. A., 2011. A performance-based system for the long-term management of municipal waste landfills. Waste Management, 31: 649- 662.

Mota C., Head M. A., Ridenoure J. A., Cheng J. J., de los Reyes III F. L., 2005. Effects of aeration cycles on nitrifying bacterial populations and nitrogen removal in intermittently aerated reactors. Applied and Environmental Microbiology, 71(12): 8565-8572.

Nathanson J. A., 2003. Basic environmental technology: water supply, waste management, and pollution control (Fourth edition). PEARSON Press, 333-340.

Niskanen A., Manfredi M., Christensen T. H., Anderson R., 2009. Environmental assessment of Ämmässuo Landfill (Finland) by means of LCA-modelling (EASEWASTE). Waste Management and Research, 00: 1–9.

Obaja D., Mace S., Mata-Alvarez J., 2005. Biological nutrient removal by a sequencing batch reactor (SBR) using an internal organic carbon source in digested piggery wastewater. Bioresource Technology, 96: 7–14.

Okereke V. O., 2002. A coupled model for evaluating the economic feasibility of leachate recirculation, In: Proceedings of the 7th Annual SWANA's Landfill Symposium, Louisville, June 17-19, Kentucky, U.S.

Pelkonen M., Kotro M., Rintala J., 1999. Biological nitrogen removal from landfill leachate: a pilot-scale study. Waste Management and Research, 17: 493-497.

Pelkonen M., Kotro M., Wang Z., 2000. Full-scale performance of biological leachate treatment at low temperature. 1st International Landfill Research Symposium, Luleå Sweden

Peng D. C., Wang Y., Wang H., Wang X. C., 2004. Biological denitrification in a sequencing batch reactor. Water Science and Technology, 50(10): 67–72.

Peng Y. Z., Zhu G. B., 2006. Biological nitrogen removal with nitrification and denitrification via nitrite pathway. Applied Microbiology and Biotechnology, 73: 15–26.

Picioreanu C., van Loosdrecht M. C. M., Heijnen J. J., 1997. Modelling of the effect of oxygen concentration on nitrite accumulation in a biofilm airlift suspension reactor. Water Science and Technology, 36(1): 47-156.

Plosz, B. Jantsch, T. G. Langford, K., Vogelsang, C., 2009. Factors influencing biological nitrogen removal in landfill leachate treatment. Proceedings: Xenobiotics in the Urban Water Cycle – XENOWAC 2009, Cyprus March 11-13, 2009. 6p.

Pochana K., Keller J., 1999. Study of factors affecting simultaneous nitrification and denitrifecation, Water Science and Technology, 6: 61-68.

Price G. A., Barlaz M. A., Hater G. R., 2003. Nitrogen management in bioreactor landfills. Waste Management, 23: 675-688.

Puig S., Vives M. T., Corominas L., Balaguer M. D., Colprim J., 2004. Wastewater nitrogen removal in SBRs, applying a step-feed strategy: from Lab-scale to Pilot-plant Operation, Water Science and Technology, 50: 89–96.

Puznava N., Payraudeau M., Thornberg D., 2001. Simultaneous nitrification and denitrification in biofilters with real time aeration control. Water Science and Technology, 1: 269-276.

Pynaert K., Sprengers R., Laenen J., Verstraete W., 2002. Oxygen-limited nitrification and denitrification in a lab-scale rotating biological contactor. Environmental Technology, 23(3): 353-362.

Rees J. F., 1980. Optimisation of methane production and refuse decomposition in landfills by temperature control. Journal of Chemical Technology and Biotechnology, 30: 458-465.

Reichel T., Ivanova L., Beaven R., Haarstrick A., 2007. Modeling decomposition of MSW in a consolidating anaerobic reactor. Environmental Engineering Science, 24: 1072-1083.

Reinhart D. R., 1996. Full-scale experiences with leachate recirculating landfills: case studies. Waste Management and Research, 14: 347-365.

Reinhart D. R., McCreanor P. T., Townsend T., 2002. The bioreactor: Its status and future. Waste Management and Research, 20: 172-186.

Robinson H., 2007. The composition of leachates from very large landfills: an international review. CWRM, 8(1): 19-32.

Ruiz C., Torrijos M., Sousbie P., Martinez J. L. and Moletta R., 2001. The anaerobic SBR process: basic principles for design and automation. Water Science and Technology, 43(3): 201–208.

Ruiz G., Jeison D., Chamy R., 2003. Nitrification with high nitrite accumulation for the treatment of wastewater with high ammonia concentration, Water Research, 37: 1371–1377.

Ruiz G. D., Jeison O., Rubilar G., Ciudad G., Chamy R., 2006. Nitrification–denitrification via nitrite accumulation for nitrogen removal from wastewaters. Bioresource Technology, 97: 330-335.

Sanphoti N., Towprayoon S., Chaiprasert P., Nopharatana A., 2006. The effects of leachate recirculation with supplemental water addition on methane production and waste decomposition in a simulated tropical landfill. Journal of Environmental Management, 81: 27-35.

SAS Institute, 2001. StatView for Windows, Version 5.0, Cary, NC, USA.

Scharff H., 2011. Sustainable sanitary landfill celebrates its 80th anniversary. Waste Management and Research, 29(8): 888.

SFS Finnish Standards Association, 1988. Determination of Chemical Oxygen Demand (COD_{Cr}) in water with the closed tube method, oxidation with dichromate. SFS 5504.

SFS Finnish Standards Association, 1990. Determination of total residue and total fixed residue in water, sludge and sediment. SFS 3008.

SFS Finnish Standards Association, 1996. Water quality. Determination of suspended solids, method by filtration through glass fibre filters. SFS-EN 872.

SFS Finnish Standards Association, 1997. Water quality. Determination of nitrite nitrogen and nitrate nitrogen and the sum of both by flow analysis (CFA and FIA) and spectrometric detection. SFS-EN 13395.

SFS Finnish Standards Association, 2004. Water quality. Determination of phosphorus, ammonium molybdate spectrometric method. SFS-EN 6878.

SFS Finnish Standards Association, 2005. Water quality. Determination of orthophosphate and total phosphorus contents by flow analysis (CFA and FIA). Part 1: method by Flow Injection Analysis (FIA). SFS-EN 15681-1.

Siegrist H., Vogt D., Garcia-Heras J., Gujer W., 2002. Mathematical model for meso- and thermophilic anaerobic sewage sludge digestion. Environmental Science and Technology, 36(5): 1113-1123.

Spagni A., Marcili-Libelli S., 2008. Nitrogen removal via nitrite in a sequencing batch reactor treating sanitary landfill leachate. Bioresource Technology. 100(2): 609-614.

Spengel D. B., Dzombak D. A., 1991. Treatment of landfill leachate with rotating biological contractors: beach-scale experiments. Research Journal of the Water Pollution Control Federation, 63(7): 971-981.

Sponza D. T., Ağdağ O. N., 2004. Impact of leachate recirculation and recirculation volume on stabilization of municipal solid wastes in simulated anaerobic bioreactors. Process Biochemistry, 39: 2157-2165.

Sormunen K., Ettala M., Rintala J., 2008. Detailed internal characterization of two finished landfills by waste sampling. Waste Management, 28: 151-163.

Statistics Finland, 2005. Finland in Figures. WWW pages on Yhdyskuntajätteet (Municipal Waste) (http://www.tilastokeskus.fi/til/jate/2004/jate_2004_2005-12-13_tau_001.html).

Stegmann R., Heyer K-U., Hupe K., Willand A., 2006. Deponienachsorge – Handlungsoptionen, Dauer, Kosten und quantitative Kriterien fuer die Entlassung aus der Nachsorge. Ufoplan Bericht, IFAS Hamburg.

Steven I. S., Bennette D. B., Robert A. P., 2004. Carbon and nutrient removal from on-site wastewater using extended-aeration activated sludge and ion exchange. Water Environment Research, 76(5): 404-412.

Strous M., van Gerven E., Zheng P., Kuenen J. G., Jetten M. S. M., 1997. Ammonium removal from concentrated waste streams with anaerobic ammonium oxidation (Anammox) process in different reactor configurations. Water Research, 31(8):1955–1962.

Tappe, W., Laverman A., Bohland M., Braster M., Rittershaus S., Groeneweg J., van Verseveld H. W., 1999. Maintenance energy demand and starvation recovery dynamics of *Nitrosomonas europaea* and *Nitrobacter winogradsky* cultivated in a retentostat with complete biomass retention. Applied and Environmental Microbiology, 65: 2471–2477.

Teichgräber B., Schreff D., Ekkerlein C., Wilderer P. A., 2001. SBR technology in Germany – an overview. Water Science and Technology, 43(3): 323–330.

Theilen U., 1995. Behandlung von Sickerwasser aus Siedlungsabfalldeponien – Betriebsergebnisse und Kosten grosstechnischer Behandlungsanlagen. Dissertation, Universität Hannover.

Third K., Newland M., Cord-Ruwisch R., 2003. The effect of dissolved oxygen on PHB accumulation in activated sludge cultures. Biotechnology and Bioengineering, 2: 238-250.

Third K. A., Paxman J., Schmid M., Strous M., Jetten M. S., Cord-Ruwisch R., 2005. Treatment of nitrogen-rich wastewater using partial nitrification and Anammox in the CANON process. Water Science and Technology, 52(4): 47-54.

Tijhuis, L., Rekswinkel, H. G., Loosdrecht, M. C. M. & Heijnen, J. J., 1994. Dynamics of population and biofilm structure in the biofilm airlift suspension reactor for carbon and nitrogen removal. Water Science and Technology, 29: 377-384.

Toettrup H., Rogalla F., Vidal A., Harremoes P., 1994. The treatment trilogy of floating filters: from pilot to prototype to plant. Water Science and Technology, 29(10): 23-32.

van de Graaf A. A., Mulder A., de Bruijn P., Jetten M. S., Robertson L. A., Kuenen J. G., 1996. Anaerobic ammonium oxidation is a biologically mediated process. Applied and Environmental Microbiology, 61: 1246-1250.van der Star W. R. L., Abma W. R., Blommers D., Mulder J-W., Tokutomi T., Strous M., Picioreanu C., van Loosdrecht M. C. M., 2007. Startup of reactors for anoxic ammonium oxidation: Experiences from the first full-scale anammox reactor in Rotterdam. Water Research, 41: 4149-4163.

van Dongen L. G. J. M., Jetten M. S. M., van Loosdrecht M. C. M., 2001. The combined Sharon/Anammox process for treatment of ammonium rich wastewater. Water Science and Technology, 44: 153-160.

van Kempen R., Mulder J. W., UijterlInde C.A., van Loosdrecht M.C.M., 2001. Overview: full scale experience of the SHARON process for treatment of rejection water of digested sludge dewatering. Water Science and Technology, 44:145–152.

Vavilin V., Rytov S, Lokshina L., Pavlostathis S, Barlaz M., 2003. Distributed model of solid waste anaerobic digestion. Effects of leachate recirculation and pH adjustment. Biotechnology Bioengineering, 81(1): 67-73.

Wang Y., 2004. Leachate pre-treatment and gas monitoring for leachate recirculation system in Ämmässuo Landfill. MSc-thesis, Helsinki University of Technology.

Wang Y., Pelkonen M., Kaila J., 2009. Implementation of on-site pre-treatment process for landfill leachate management: a cost-savings estimation case study. In: Proceedings Sardinia 2009, Twelfth International Waste Management and Landfill Symposium S. Margherita di Pula, 5 - 9 October Cagliari, Italy.

Warith M., 2002. Bioreactor landfills: experimental and field results. Waste Management, 22(1): 7-17.

Warith M., Li X., Jin H., 2005. Bioreactor landfills: state-of-the-art review. Emirates Journal for Engineering Research, 10 (1): 1-14.

Willers H. C., Derikx P. J. L., ten Have P. J. W., Vijn T. K., 1997. Nitrification limitation in animal slurries at high temperatures. Bioresource Technology, 64: 47-54.

Wiszniowski J., Robert D., Surmacz-Gorska J., Miksch K., Malato S., Weber J-V., 2004. Solar photocatalytic degradation of humic acids as a model of organic compounds of landfill leachate in pilot plant experiments. Applied Catalysis: B Environmental, 53(2): 127-137.

Yalmaz G., Öztürk I., 2001. Biological Ammonia Removal from Anaerobically Pre-treated Landfill Leachate in Sequencing Batch Reactors (SBR). Water Science and Technology, 43: 307-314.

Yalmaz G., Öztürk I., 2003. Nutrient removal of ammonia rich effluents in a sequencing batch reactor, Water Science and Technology, 48: 377-383.

YTV, 2010. Jätteenkäsittelykeskuksen toiminta vuonna 2009 (Waste management activities in 2009). WWW pages on HYS (http://www.hsy.fi/jatehuolto/ymparisto).

Yuen S. T. S., 2001. Bioreactor landfills: Do they work? Geo-environment 2001: 2nd ANZ Conference on Environmental Geotechnics, Newcastle, Australia, November, 28-30.

Zeghal S., Nagem Nogueira F., Salzer C., Rogalla F., 1995. Tertiary denitrification using an upflow floating biofilter. WEFTEC 68th Annual Conference and Exposition, Miami Beach, Florida, October, 21-25.

Zeghal S., Sibony J., 1996. Biofilter applications catalog for nutrient removal. WEFTEC 67th Annual Conference and Exposition, Dallas, October, 5-9.

Zehnder A. J. B., 1978. Ecology of methane formation. Water Pollution Microbiology, 2: 349-376.

Zeng R, Lemaire R, Yuan Z and Keller J., 2003. Simultaneous nitrification, denitrification and phosphorus removal in a lab-scale sequencing batch reactor. Biotechnology and Bioengineering, 84: 170-178.





ISBN 978-952-60-5140-6 ISBN 978-952-60-5141-3 (pdf) ISSN-L 1799-4934 ISSN 1799-4934 ISSN 1799-4942 (pdf)

Aalto University School of Engineering Department of Civil and Environmental Engineering www.aalto.fi BUSINESS + ECONOMY

ART + DESIGN + ARCHITECTURE

SCIENCE +

CROSSOVER

DOCTORAL DISSERTATIONS