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Kai Markus Sormunen

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UNIVERSITY OF JYVÄSKYLÄ

JYVÄSKYLÄ 2008

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ABSTRACT

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Different types of municipal solid waste (MSW) landfills were characterized according to waste and leachate sampling, on-line measurements of internal leachate and landfill gas monitoring to provide information for both active landfill and post-landfill operations in boreal conditions. The study showed that waste and internal leachate, organic and nitrogen content, and biological methane potential (BMP) as well as their distribution can to a certain extent be characterised in landfills with unknown/undetermined contents. All parameters showed high horizontal and vertical variation in each landfill indicating that both the composition and state of degradation of waste varied greatly in these landfills. For example, the BMP (range 1-183 m^3/t total solids (TS) and 6-60 m^3/t TS in the 20- and 50-years-old landfills, respectively), volatile solids (VS, range 35-89 % and 3-80 %) and leaching of ammonium-nitrogen (range 0-2205 g/t TS and 0-1400 g/t TS) were higher in the 20-year-old than 50-year-old landfill. The rates of methane generation and potential between the landfills were different, as indicated by the first order kinetic methane generation rate (*k*) and methane generation potential *(L)* for bulk waste, which, on the basis of experimentally determined methane potentials and modelling in addition to actual landfill methane recovery, were 0.17 and 130 m^3/t in the 20-year-old and 0.05 and 40 m^3/t in the 50-year-old landfills, respectively. The mechanically and mechanically-biologically treated MSW residuals were characterized by different nitrogen and organic matter budgets as well as different leachate and gas properties. The mechanically treated MSW residuals prolonged by at least two years the acidogenic phase of degradation (pH 5.8-6.1) and leaching of ammonium nitrogen (391 g/t TS; 8 % of initial nitrogen content) and organic matter (24 kg COD /t TS), while with biologically treated residuals the methanogenic phase of degradation was reached (pH >7) after a few months and the leaching of ammonium nitrogen (79 g/t TS; 1 % of initial content) and organic matter (0.6 kg/t TS) were significantly lower during the 14-month study period.

Key words: Landfill; characterisation; municipal solid waste; methane; nitrogen; monitoring; on-line.

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CONTENTS

LIST OF ORIGINAL PUBLICATIONS

The thesis is a summary and discussion based on the following articles and manuscripts, which are referred to by their Roman numerals I - V in the text.

I planned the studies together with my co-authors, did the major part of the experimental work, and, with the support of my co-authors, processed the data. I also wrote the first drafts and revised them after comments by my co-authors.

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

II Sormunen, K., Ettala, M. & Rintala, J. Internal leachate quality in a municipal solid waste landfill: vertical, horizontal and temporal variation and impacts of leachate recirculation. (Accepted tentatively)

III Sormunen, K., Ettala, M. & Rintala, J. Long-term on line monitoring of water table and leachate quality in a municipal solid waste landfill. (Manuscript)

IV Sormunen, K. & Rintala, J. Determination of first order kinetic methane generation factors (rate *k* and potential *L)* and methane generation potentials for two Finnish municipal solid waste landfills. (Submitted)

V Sormunen, K., Einola, J., Ettala, M. & Rintala, J. 2007. Leachate and gaseous emissions from initial phases of landfilling mechanically and mechanically-biologically treated municipal solid waste residuals. Bioresource Technology. (In press)

ABBREVIATIONS

BMP	biological methane potential
BOD	biological oxygen demand
COD	chemical oxygen demand
EU	European Union
EC	European Commission
GB21	gas production during 21 days
GSM	global system for mobile communications
ha	hectare
IPCC	intergovernmental panel on climate change
k	methane generation rate
L_o	methane generation potential
L/S	liquid / solid
M	mechanical
MAP/PET	mean annual precipitation / potential evapotranspiration
MB	mechanical-biological
MSW	municipal solid waste
TKN	total kjeldahl nitrogen
NH_4-N	ammonium nitrogen
REF	recycled fuel
SCOD	soluble chemical oxygen demand
TS	total solids
USEPA	United States Environmental Protection Agency
VFA	volatile fatty acids
VS	volatile solids

1 INTRODUCTION

1.1 Trends in municipal solid waste landfilling

A large variety of discarded materials has always been generated. Early on, the small amounts of discarded material generated were mainly organic, biologically decomposable and dispersed over large areas, and thus major problems with waste materials did not arise. Late, in the course of twentieth century, along with increased trading changes in the material industry (e.g., plastics, inorganic and forest industry materials) led to changes in the composition and properties of discarded materials. The most significant change was probably the commercial production of plastics which combined with increase in the size of population and consumption of materials. Since then, increasing amounts and kinds of materials have been produced and, after use, discarded as an unwanted material - waste. Commonly, waste was locally dumped along with surplus soil and rock and other low value materials. However, the increasing volumes of waste in dumps produced nuisances such as odours and flies. In response the use of daily covers (soils) and the concept of sanitary landfilling were developed in the 1930's (CEC 1992). Since, compaction, modern filling methods, trenching, drainage and leachate collection and landfill gas recovery systems have been introduced. In spite of the technologies developed and increasingly applied, gaseous (e.g., methane as a greenhouse gas) and leachate (e.g., organic matter and nitrogen) emissions continue to occur during landfilling and thus a lot of effort has been focused on the structural (e.g., liners, cover layers for methane oxidation) and operational (aeration, leachate recirculation) management of landfills or pre-treatment of waste before landfilling in order to minimize areas needed for landfills and to reduce emissions into the environment.

At the end of the twentieth century new waste management strategies and treatment methods were introduced, including source segregation of different waste materials, composting, anaerobic digestion, mechanical and mechanical-biological treatment as well as use of recycled fuel (REF) or modern incineration.

Moreover, recycling and waste reduction strategies of various kinds have been considered important prior to landfilling to minimize air and water pollution and to save natural resources. These measures are especially important, because of the increasing numbers of new landfills that have been established along with urbanisation, industrialisation and rising standards of living in the developing and newly industrialized countries.

Landfilling has continued to be a major method of municipal solid waste (MSW) disposal during recent years (Eurostat 2005, USEPA 2005a). For example, in the European Union (EU) approximately 49 % (118.5 million tonnes) of all municipal waste (total 243 million tonnes) was landfilled, 17 % incinerated and 34 % recycled or treated otherwise in 2003 (Eurostat 2005). Since 2000 landfilling in the EU has slightly fallen due to increased recycling and incineration following the EC Landfill Directive (1999/31/EC).

The requirement that landfills must have a bottom liner means that a large number of landfills in the EU will be closed by 2007. The EU directive will also phase out the quantity of organic waste which can be landfilled; thus waste minimisation and pre-treatment before landfilling are being encouraged. This in turn affects the composition of waste that can be landfilled, rendering it more homogenous and stable. According to the EC directive (1999/31/EC), the share of biodegradable landfilled MSW had to be reduced by 25 % before the year 2006, compared to the amount of biodegradable waste in 1994, and will have to be reduced by at least a further 50 % before 2009 and 65 % before 2016. In some EU countries even more stringent national requirements were set: for example, in Germany it has been possible to landfill only thermally and mechanically-biologically pre-treated MSW since June 2005 (Stegmann 2005). Source segregation of biowaste (kitchen and garden waste), papers, cardboard and energy waste (e.g., plastics and non-recyclable papers such as tissues, paper cups, food containers, brown papers and binders) might have been enough in some EU countries to meet the aims for 2006, while the further requirements (from 2009 onwards) will need other methods such as mechanical, mechanical-biological treatment or incineration. From the 1990s, Finnish MSW has been increasingly segregated at source into biowaste, glass, metals, paper and cardboard and residuals. The residual fraction as such or after the mechanical removal of materials for recycled fuel (REF) has commonly been landfilled, and thus the amounts of landfilled biodegradable materials have generally fallen in Finland. This in turn may minimize landfill gas production and the leaching of organic materials and nitrogen. However the implementation of source segregation continues to vary greatly between local municipalities, which probably causes regional differences between landfills. Differences in landfilled waste characteristics may therefore affect landfill gas collection, treatment and utilization, or other operations (e.g., leachate recirculation), especially during the post-closure of landfills.

Alongside waste treatment technologies landfill bioreactors have been introduced in order to both control and promote degradation and gas production in MSW landfills (e.g., Benson et al. 2007, Yazdani et al. 2006). Traditionally, the main operation in landfill bioreactors has been leachate recirculation; however, within the last decade aeration of landfills has also been developed (Heyer et al.

2005). Landfill bioreactors have been developed for the treatment of untreated MSW, while only a few studies exist on bioreactor operation by leachate rerirculation with mechanically (Woelders & Oonk 1999) and mechanically-biologically (Lorber et al. 2001) treated waste.

To summarize, landfilling will remain an active part of the waste management. Recently, increased attention globally has been paid to the characteristics of landfilled waste and control of degradation or conditions in the landfill body with aim of seeking to minimize the environmental impacts of landfills.

1.2 Waste characterisation and stages of degradation in landfills

The quality of landfilled MSW has varied along with the practises of industrialization, consumption and waste management. Recently, the major fractions (Table 1) of discarded MSW have been paper and cardboard, kitchen biowaste, plastics and garden waste (Golder Associates 1999, USEPA 2005c, YTV 2004), and major portions of the methane potential can be attributed to cellulose and hemicellulose (Baldwin et al. 1998, Barlaz et al. 1989). The emission (and energy) potential of different MSW fractions vary greatly; e.g., the source segregated residual fraction of MSW (termed "grey waste" in Finland) and biowaste may have a biological methane potential (BMP) of 46 m^3/t total solids (TS) (grey waste) and 410 m^3/t TS (biowaste) and contain 2.1 kg NH_4-N/t TS (grey waste) and 3.6 kg NH_4-N/t TS (biowaste) of leachable nitrogen (Jokela et al. 2002). Furthermore, in addition to waste, landfills often contain soil of variable properties which is used as daily cover.

TABLE 1 Composition of MSW (wet weight) after source segregation of recyclable materials in Finland, U.S. and Australia (I).

Waste	Finland (%)[1]	U.S. (%)[2]	Australia (%)[3]
Paper and paper/cardboard	20	26.3	9.9
Glass	4	6.2	6.8
Metals	4	7.3	7.1
Plastics	13.6	15.4	7.3
Rubber and leather	--	3.5	--
Textiles	4	5.5	--
Kitchen biowaste	38 (including garden waste)	16.4	38.1
Garden waste	--	7.6	17.8
Wood	3	7.5	6.4
Inorganic Wastes	--	2.2	--
Diapers	7	--	--
Other combustible	3	--	--
Other	3.4	2	6.6

[1]YTV 2004; [2]USEPA 2005c; [3]Golder Associates 1999

In landfilling the waste is compacted to achieve a density varying from 0.8 t/m^3 to 1.0 t/m^3 (wet weight) for untreated municipal waste (CEC 1992). The height of landfill bodies generally range from a few meters up to >100 m and landfill areas from a few hectares as much as 40-50 hectares. Landfills are actively filled for several decades or, if separate cells are used, for a just few years.

Thus waste age, properties and rates of degradation are likely to vary both horizontally and vertically. Due to biodegradation and the gravitational effects of overlying waste layers, settlements, typically from 5 to 20 % and even up to 35 % in respect to landfill height, may occur (CEC 1992). After a settling period of few years landfills are closed by cover layers to limit the infiltration of rainwater and to minimize leachate generation and gaseous emissions. However, limiting infiltration will decrease the water content and movement of leachate, which in turn will have effect on the transport of nutrients, organic materials and microbes. In general, sealed cover layers have the effect of minimizing biological activity and waste stabilization in landfills. Therefore leachate recirculation has been considered important (e.g., Benson et al. 2007, Yazdani et al. 2006) to maintain the degradation of organic materials as well as to leach out inert materials (e.g., chloride) and intermediate degradation products (e.g., NH_4-N) to obtain a higher rate of stabilization (Reinhart & Al-Youshi 1996).

In landfills waste undergoes various physical, chemical and biological reactions, all of which have an effect on the composition of landfill gas and leachate. However biological reactions play the major role in MSW landfills containing organic materials, thus regulating the rate and stages of degradation. The degradation of biodegradable waste in MSW landfills is typically identified by reference to four (Farquhar & Rovers 1973; cited in Kuehle-Weidemeier 2004) or five (Pohland & Al-Youshi 1994, Reinhart & Al-Youshi 1996) phases: aerobic, acidogenic, transition (or unsteady methanogenic), methanogenic and maturation. The first three phases may happen within three years, while the fourth phase may continue for several decades (Bockreis & Steinberg 2005). Up to five separate phases have been further identified in the maturation phase (Franzius 1981, Rettenberger & Metzger 1992; cited in Kuehle-Weidemeier 2004). The existence of these additional phases have not been characterised in landfill conditions due to fact that methane phase continues to prevail in landfills under post-closure operation. The acidogenic phase seems especially to promote the leaching of organic materials as high amounts of volatile fatty acids (VFA) are produced. These may decrease pH to a level non-optimal for methanogenic bacteria. Low populations of methanogenic bacteria in turn are unable to utilize these acids at same rate as they are produced in fresh waste (Barlaz et al. 1990). The gas generated in the acidogenic phase is mainly carbon dioxide and hydrogen, while in the transition phase methane generation starts (Bockreis & Steinberg 2005). In the transition phase VFAs are increasingly consumed by the methanogenic bacteria in the landfill and pH increases towards the level optimal (6.8-7.4) for methane production, thereby reducing the leaching of organic materials (Barlaz et al. 1990).

In practice the identification of acidogenic and methanogenic conditions has been considered important as these phases seem to have markedly different

leachate (Ehrig 1983, Kjeldsen et al. 2002) and gas characteristics (Barlaz et al. 1990, Bockreis & Steinberg 2005, Pohland & Al-Youshi 1994, Reinhart & Al-Youshi 1996), which in turn determine the requirements for leachate and gas treatment and the possibility for methane utilization. Moreover, the short (up to few days, Kjeldsen et al. 2002) aerobic phase seems to decrease the content of organic materials (measured as COD) in leachate (Ehrig 1983). However the duration of aerobic phase is often curtailed due to the use of a temporary cover layer or intensive waste filling.

It has been noted that climatic conditions such as precipitation and temperature affect the degradation of landfilled waste (IPCC 2006). In boreal conditions, as in Finland, the temperature may vary from -40 to 30 °C in the same areas while the mean temperature remains below 0 °C for 4 to 7 months in the country as a whole with the precipitation varying from 500 to 650 mm (Finnish Meteorological Institute 2007). Thus the rate of degradation is probably less than rate in the warmer climatic conditions with higher precipitation. Previous study (Garg et al. 2006) concluded that the amount of precipitation has a more important effect on waste degradation than temperature. However, in Finland a big proportion of the annual precipitation is typically in the form of snow, which minimises infiltration in the winter months, while melting waters may cause high infiltration in the spring. Thus varying seasonal climatic conditions may affect the characteristics of landfills in boreal conditions.

1.3 Operation and monitoring of landfills

Stated as above, the need to minimise the environmental impact and to optimise land use has resulted in new waste management strategies. These new strategies aim to control landfill waste quality or develop methods of operating active or closed landfill to obtain a faster rate of stabilization. Monitoring both the functionality of landfill structures and stages of waste degradation have in turn resulted in a need for new monitoring methods in landfills in which the waste characteristics are not precisely known and which have highly heterogeneous materials and varying conditions. In general, the number of unkown properties causes uncertainty when seeking to accelerate degradation or minimize emissions through ways such as leachate recirculation, aeration and or methane oxidation. Thus determination of the methane generation potential of a landfill is an important step before implementing gas recovery and the utilisation of gas in energy production or sizing the biological active cover layer for methane oxidation. Moreover the degree of stabilization with regard to organic materials and nitrogen have rarely been determined by waste sampling and analysis, even if assumptions about the length of post-closure periods have probably been made for most landfills under operation and post-closure monitoring.

Landfills with unknown MSW content have previously been sampled to estimate the rate of degradation of MSW and its different components (e.g., Baldwin et al. 1998, Bogner 1990, Gardner et al. 2003, Gurijala & Suflita 1993, Hartz & Ham 1983, Jokela et al. 2002) while only a few studies have been published on vertical profiles pertaining to pH, temperature, moisture, organics, cellulose, lignin, or BMP (Attal et al. 1992, Bookter & Ham 1982, Chen et al. 2004, Ham et al. 1993, Jones et al. 1983, Östman et al. 2006, Townsend et al. 1996, Wang et al. 1994) and even fewer studies (Ettala et al. 1988, Ham et al. 1993, Östman et al. 2006) on landfill nitrogen content. These earlier studies showed MSW landfills to be heterogeneous with respect to stages of degradation and conditions within the landfill body, with wastes in the top layers usually less degraded than those in the deeper layers.

Recently, landfills have been monitored in order to determine their mechanical stability (settling), water table, temperature, gas and leachate generation as well as such characteristics as landfill methane content and the amounts of nitrogen and organic matter in leachate. Typically these have all been monitored a few times in a year in the case of settling and leachate characteristics or continuously in the case of the volume of gas recovered and leachate generated. On the basis of these measurements it may have been possible to draw some conclusions about the conditions and stages of degradation of landfills and functionality of environmental protection systems. However some measurements, which have generally been considered important, such as methane emissions (IPCC 2006) together with gas recovery efficiency have been rather neglected due to limitations in the methane emission monitoring technologies available.

Monitoring of the water table and temperature has been considered important, especially when operating a landfill as a bioreactor, due to fact that a rise in the water table may cause increasing leaching into the groundwater and a high temperature can damage the lining as well as leachate and gas collection systems (Benson et al. 2007). Also, according to Finnish legislation (Finnish Government 1997), it is important to monitor the water table in the internal leachate. Moreover, temperature monitoring is done in order to determine landfill conditions and biological activity. However, on-line monitoring is often not thought to be necessary, despite the fact that, as stated in the legislation (Finnish Government 1997), it is necessary to ensure that the various processes of degradation proceed as intended and that environmental protection structures (e.g., drainage layer, liner and leachate recirculation structures) are fully functional. Moreover it is important that monitoring is systematic, samples are representative and that changes in leachate quality are noted quickly. Changes in the water table and leachate quality have traditionally been monitored by sampling and laboratory analysis or field measurements, while internal on-line monitoring has rarely been studied, even if on-line monitoring (e.g., pH, electrical conductivity, COD, turbidity) has been widely used in wastewater quality monitoring (Thomas & Pouet 2005).

2 OBJECTIVES

The main objective of this study was to evaluate the feasibility of different methods of characterizing MSW landfills and to characterize different kinds of landfills in boreal conditions so as to obtain information of relevance for both active landfill and post-landfill operations.

The specific objectives were:

- To determine the horizontal and vertical variation in waste and leachate properties as well as temporal variation in the water table, temperature and leachate quality in old MSW landfills with unknown contents.
- To study the kinetic factors (methane generation rate (k), methane potentials (L_o)) in old MSW landfills on the basis of their waste properties and gas recovery data so as to be able to estimate the methane generation potential and present recovery rates.
- To study on-line monitoring of the water table, temperature, electrical conductivity and pH in the internal leachate quality, assess the technical feasibility of these measurements, and evaluate the utility of the data obtained.
- To determine gaseous and leachate emissions and to evaluate the feasibility of the nitrogen and organic matter budget in order to characterise the landfilling of mechanically and mechanically-biologically treated MSW residuals with known contents.

3 MATERIALS AND METHODS

3.1 Study areas

3.1.1 Ämmässuo landfill (I, III, IV)

In Ämmässuo landfill (the largest landfill in Scandinavia, established in 1987 in metropolitan Helsinki), the total amount of landfilled waste is about 8 million tons. Waste is currently being deposited at a current annual rate of 0.5 million tons, of which 0.3-0.4 million tons is MSW and the rest mainly industrial and construction waste (Table 2, Karhu 2004). Initially MSW was totally landfilled; however, during the 1990s source segregation (biowaste, paper and cardboard, glass and metals) gradually increased in this region and from the end of that decade, only the residual fraction of MSW (grey waste) has been landfilled. Historically, MSW has been disposed in three sectors of this site (sectors referred as 1, 2 and 3, Fig. 1) so that a similar waste history can be assumed across the landfill. Waste was compacted in horizontal layers with soils, composted sewage sludge or composted biowaste as daily cover. For this study samples from sector 3 (40 samples) and sector 1 (4 samples) were taken: in sector 3 from two to four depths at 15 boreholes (Ø 1.2 m) and in sector 1 at 4 boreholes (one sample per borehole) by grab sampling during the installation of the gas collection wells using a drilling rig. Waste materials were crushed during drilling so that the largest particles were typically <20 cm in diameter. Larger fragments of metals, plastics, and stones which were not crushed were removed and weighed during sampling. The weight of all removed materials was included in the results. The samples were taken from +62 to +82 m above mean sea level; the bottom of the waste was +60 to +62 m above mean sea level. The temporary cover of the landfill varied from +82 to +94 m at the sampling points, hence the sampling levels were reported from mean sea level.

Two separate systems based on gravitional and pressurised recirculation were installed in 2002. Both leachate recirculation areas (0.5 ha per area) were located at a distance of 10 to 15 m from each other and leachate recirculation

was practised in both areas from June to October in 2003 and 2004. The volume of recirculated leachate was approximately 310 mm in 2003 and 460 mm in 2004 in the area with the pressurised system, while in the area with the gravitional system the corresponding values were 140 and 160 mm.

The monitoring wells used in present study for on-line monitoring of water table and temperature were supplied around the study area based on pressurised leachate recirculation. Four instrumented monitoring wells (named A, B, C, D; depths between 8.5 and 21.5 m, Table 3) for temperature and water table monitoring were bored in 2003. Instrumented monitoring wells were bored on the landfill slope, where height of the landfill varied from 10 to 24 meters. Monitoring wells A, B and D were in close proximity to the leachate recirculation canals, the distance from a monitoring well to the closest leachate recirculation canal varying from 5 to 18 m, while monitoring well C was located at approximately 80 m distance from the leachate recirculation area.

The monitoring wells were instrumented by a piezoresistive pressure transmitter (Keller PR-36W) for the water table and a thermistor (Betatherm 10K3A1B) for temperature. All the monitoring wells extended from 2 to 4 m from the bottom liner of the landfill. The monitoring wells were equipped with screens varying from 2 to 13 m and extensions from 4 to 22 m. Data were recorded hourly (for periods between 194 and 609 days during the study from 10.4.2003 to 29.11.2004) in a datalogger (Cambell CR10X) and transferred by a GSM modem (Siemens TC35i) with datalogger support software (Campbell Scientific, version PC208W) to a computer as required. The datalogger, GSM modem, accessories and battery were located in a mast-mounted measuring station on the landfill. The datalogger and GSM modem were powered by a solar photovoltaics panel installed outside the measuring station.

TABLE 2 Characterisation of Ämmässuo and Kujala landfills (I).

Parameter	Ämmässuo landfill	Kujala landfill
Established in	1987	1955
Landfilled waste (1000 t/a)	500 – 600	20 – 70
Area (ha)	54	27
Waste amount (million t wet)	7.4 (in 2004)	3.2 (in 2003)
Altitude (m from sea level)		
Bottom structure	+60 - +62	+99 - +100
Max. altitude (at sampling	+94 in 2004	+123 in 2003
points)	+68 - +72 in 2003	+104 - +110 in 2003
Water table		
Start of source segregation :		
Biowaste	1995[1]	1994[1]
Energy waste		1998[1]
Paper and cardboard	during 1990s[2]	1998[1]
Other (e.g., glass and metals)	areal collection 1990s	areal collection 1990s
Annual Rainfall (mm)	601-700[3]	601-700[3]
Leachate recirculation or irriga-	not used in sampling area	irrigation in sampling area in
tion		1970-2000
Leachate formation (m^3/a)	85 775 (in 2000)[4]	55 785 (in 2003)[5]
Leachate characteristics[4]:		
NH_4-N (mg/l)	570	43-210 (mean 112)
N_{tot} (mg/l)	580	62-222 (mean 126)
COD_{cr} (mg/l)	1600	250-550 (mean 367)
TOC (mg/l)	--	110-170 (mean 140)
BOD_7 (mg/l)	210	80[4] (one analysis)
pH (in observation tubes, own	6.0-7.5	6.8-8.2
data, not shown)		

[1] Residential buildings containing more than 10 apartments; [2] Residential buildings containing more than 20 apartments; [3] Finnish Meteorological Institute 2007; [4] Annual mean values and/or ranges measured from regular monitoring of leachate quality (in Ämmässuo: Maa ja Vesi Oy 2002, and in Kujala: Vääränen & Tuominen 2004, Vääränen 2005)

TABLE 3 Height of landfills in on-line instrumentation locations, and bottom as well as top levels of monitoring wells with their screens in Ämmässuo (III).

Ämmässuo	Height of landfill (m)	Depth of monitoring well (m)	Screen range above mean sea level[1] (m)	Lengths of screen (m)	Distance from closest recirculation canal (m)
A	24	21.5	64.4-66.4	2	15
B	10	8.5	62.9-68.9	6	18
C	20	16	63.6-65.6	2	80
D	17	16	61.9-74.9	13	5

[1] Screen range measured from mean sea level

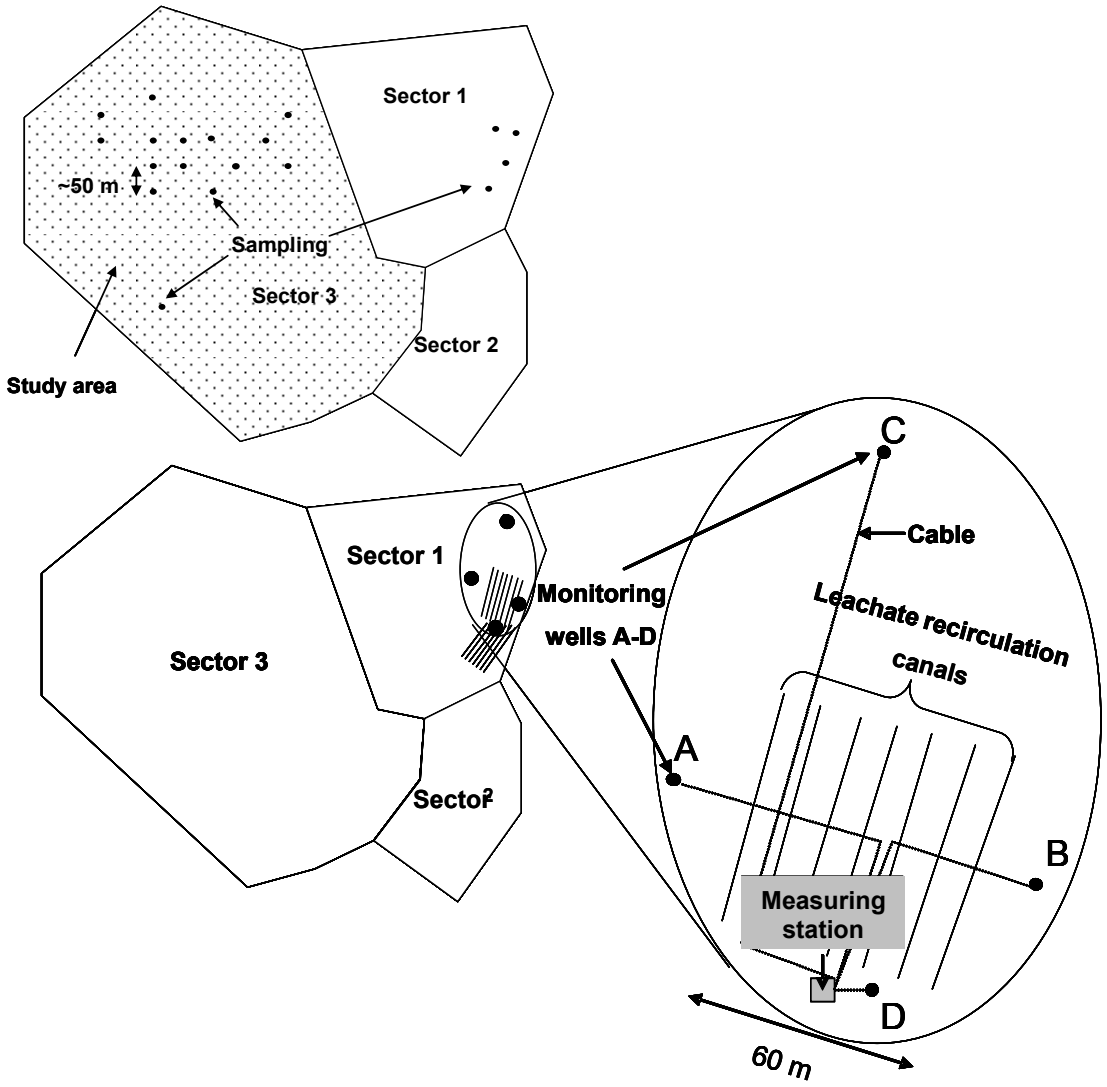

FIGURE 1 Location of waste sampling (top) and on-line monitoring points in Ämmässuo landfill (I, III).

3.1.2 Kujala landfill (I, II, III, IV)

The Kujala landfill (Lahti, Finland) was initially established for surplus soils and MSW (Ettala et al. 1988). The amount of landfilled waste was approx. 3.2 million tons in 2003. From 1965 to 1984 the landfilled material consisted mainly of incineration residuals of MSW. In 1965 approx. 200 tons of MSW and in 1975 – 1984 over 20 000 tons of MSW was incinerated. Sewage sludges were landfilled until 1980. Thereafter the rate of landfilling was increased from 20 000 (in 1981) to 67 000 tons (in 2004) of which about 44 000 tons were classified as MSW while the rest was industrial (ash, wood dust) and construction waste (PHJ 2005). MSW was landfilled from 1984 until 1994 when biowaste source segregation was implemented. The landfilled waste underwent further changes after 1998 when source segregation of MSW into biowaste, energy waste, paper, cardboard, metal, glass and landfill waste was introduced. Waste is landfilled in

horizontal layers and compacted by a landfill compactor using soil for daily cover. Waste was commonly burnt on landfill site from 1960s until the 1980s; in addition, leachate recirculation via sprinklers was practiced during the summer from the 1970s to the 1990s. The bottom level of the landfill (no liner) is at +99 to + 100 m above mean sea level and the temporary cover layer was at +115 to + 123 m above mean sea level at the time of this study (in 2003). The waste in the study area was landfilled from the 1960s to 2003.

Fourteen monitoring wells (inner diameter 50 mm) were installed of which ten (C1, C2, D1, E1, E2, F1, G1, G2, C3 and G3) were located in the leachate recirculation area at a distance of approximately 5-25 m from the leachate recirculation canals, while four monitoring wells (A1, B1, H1, H2) were located in the control area at approximately 25-60 m from the leachate recirculation canals in Kujala landfill (Fig. 2, Table 4). The leachate recirculation was practiced from June to October (approximately 600 mm in 2003 and 500 mm in 2004) by two horizontal leachate recirculation lines (length 200 m) located at depth of approximately 3 meters from the temporal cover layer. The monitoring wells were horizontally located at intervals of 5 to 100 meters and vertically located so that the bottom of the monitoring wells were situated at varying levels up to 6 meters from the bottom. The 11 monitoring wells were equipped with a long screen (varying from 6.5 to 21 m) with a short (3-4 m) extension leading to the top of the landfill. Wells C2, E2 and G2 (with varying screen lengths from 16 to 18 meters) were installed in parallel (distance 5 meters) with wells C1, E1, G1 (screen length 2 meters) to obtain samples from the upper leachate at 3.5 to 6 m distance and close (< 1 m) to the bottom of the landfill.

For the on-line measurement of the water table and temperature in the monitoring wells a combined sensor (Labko 4390-027) for temperature and water table was installed in four monitoring wells (H1, G2, G3 and C3). Moreover on-line conductivity was studied in three monitoring wells (C1, G3 and H1; electrode GLI 3725E) and pH in one monitoring well (H1; electrode GLI PD1P1). The monitoring wells were equipped with screens between 2 and 21 m and extensions from 2 to 23 m. The water table and temperature were monitored for varying periods between 212 and 516 days and conductivity (C1, G3 and H1) and pH (H1) for 167 days. Data transfer from electrodes (through logic control and a radiomodem) to the control room was continuous, and real-time data was displayed both on a field monitor located in the proximity to monitoring wells and in a monitor located in the control room. On-line data were saved once per hour in the control room computer. The measuring systems on the landfill were connected to an electrical network.

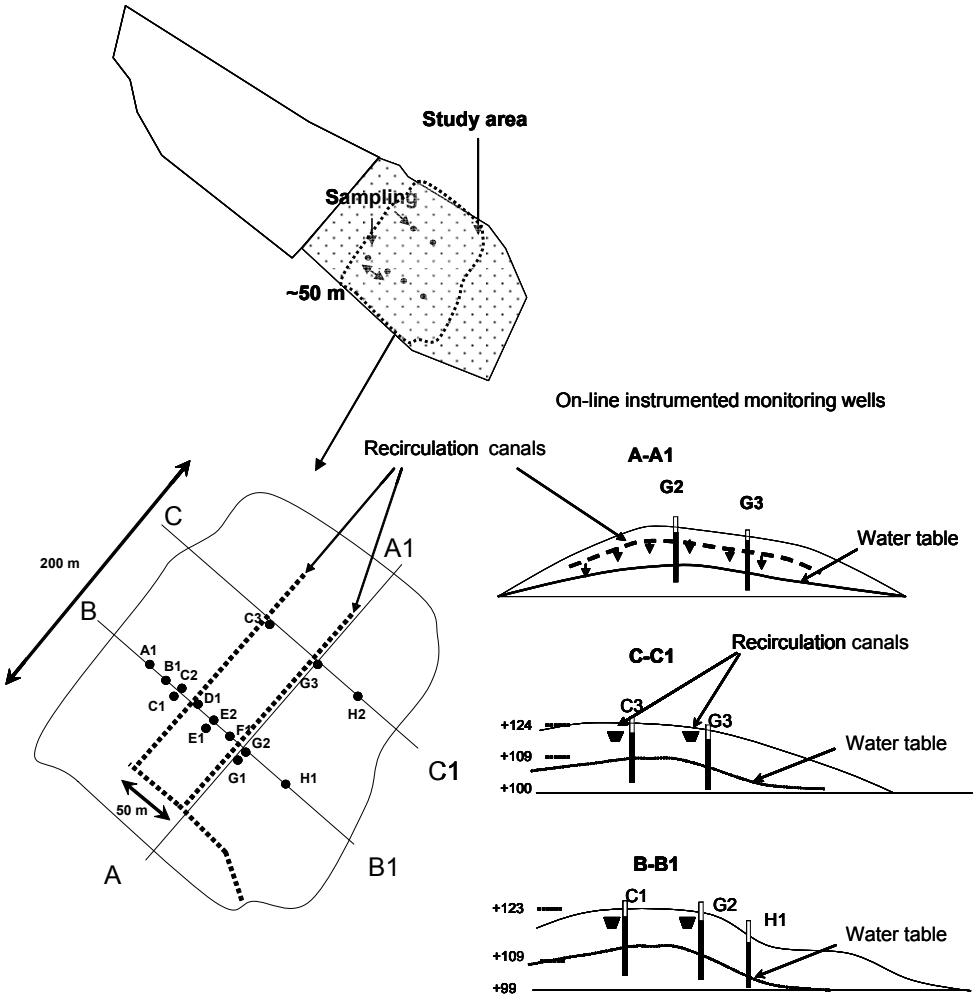

FIGURE 2 Study area and location of waste sampling points (top), internal leachate monitoring wells (A1, B1, C1…; bottom, left) and on-line instrumented monitoring wells (C1, C3, G2, G3, H1; bottom, right) in Kujala landfill (I, II, III).

3.1.3 Landfill lysimeters (V)

Leachate and gaseous emissions during the initial phases of landfilling mechanically (M) and mechanically-biologically (MB) treated municipal solid waste residuals were characterised using two landfill lysimeters. The lysimeter were furnished with systems for measuring leachate flow and temperature with facilities for leachate and gas sampling. The lysimeters were monitored over two years. The lysimeters (height 3.9 m, width 2.4 m, length 12 m, volume 112 m^3) made from steel frames (RHS 60 * 80 mm) and walls (2 mm) and coated with acryl paint (Hempatex Hi-build 46410) were placed in a 30 years old waste and soil landfill body in November 2003 (Fig. 3).

TABLE 4 Landfill height at locations of monitoring wells, screen range of monitoring wells and distance from leachate recirculation canals (II).

Monitoring well	Height of landfill (m)	Height of monitoring well (m)	Screen range above mean sea level (m)	Distance from closest recirculation canals (m)
A1	22.4	19.0	102.4-119.4	45
B1	24.8	21.0	102.0-120.0	25
C1	24.6	25.0	99.9-101.9	5
C2	25.1	21.0	104.1-122.1	5
C3	25.0	24.0	100.5-121.5	5
D1	25.3	22.0	103.3-122.3	5
E1	24.4	25.0	99.5-101.5	25
E2	24.6	19.7	104.9-122.9	25
F1	22.5	19.4	103.1-119.1	5
G1	21.5	23.5	98.7-100.7	5
G2	21.5	19.0	102.5-118.5	5
G3	18.2	16.7	101.5-115.2	5
H1	13.2	13.3	99.9-109.9	55
H2	8.6	9.5	100.8-107.3	60

The lysimeters were filled with the mechanically (M) and the (mechanically-biologically) MB residuals in 0.5 m horizontal layers, which were compacted by a soil compactor (Bomag 105, 1.6 t). The densities obtained were 0.9 and 1.0 t/m^3 (wet weight) in M and MB residuals, respectively (Table 5). The leachates were gravimetrically (angle 5 °) collected from the drainage layer (thickness 30 cm, gravel particle size <25 mm) and collection drain (110 mm) to flow-meter wells. The lysimeters were covered by plywood board covers during approximately the first 120 days of the study (from 1st of December 2003 to 1st of April 2004) and thus the leachate flows were low before April 2004. In June 2004 tap water (2 x 500 litres) was added to the lysimeters to promote leachate formation and to monitor the leachate flow by a tracer (lithium chloride) method (data not shown). The added water (1000 l) was counted as extra rainfall.

M residual was obtained from Loimi-Häme Regional Solid Waste management Ltd (Forssa, Finland). In the Loimi-Häme region metals are source-segregated, and biowaste and papers are source-segregated in residential buildings containing more than five households, while in the case of other buildings biowaste, paper, cardboard and glass are source-segregated where the segregated waste streams are >20 kg per week. Furthermore, a network of local collection points for papers, metals and batteries exists for households.

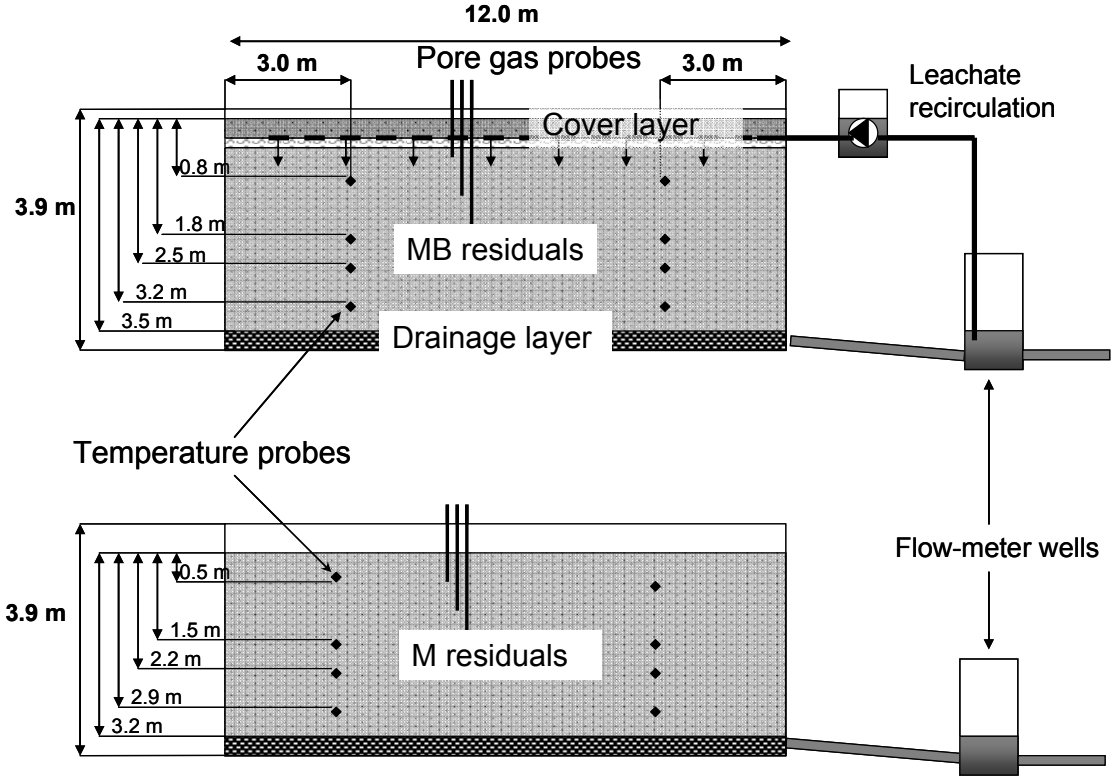

FIGURE 3 Sketch of MB (top) and M residual (bottom) in the landfill lysimeters (V).

TABLE 5 The characteristics of M and MB residuals in the landfill lysimeters (V).

Parameter	M	MB
Wet weight (t)	84	97.8
TS (%) and (t TS)	67 and 56.3	54 and 52.8
VS (%) and (t VS)	41 and 34.4 t	23 and 22.5
VS/TS (%)	61	43
pH	5.9-6.5	7.0-7.6
BMP (m^3/t TS) and (m^3/t wet)	259 and 105	52 and 21
TKN (%) and (kg)	0.5 and 412 kg	0.6 and 548 kg
Height (m)	3.2	3.5
Volume (m^3)	92	96
Density (t/m^3)	0.9	1.0 (cover layer 0.8)[1]

[1] The cover layer contained gravel 10-15 cm and 40-45 cm (9.8 t, slightly compacted) MB residual

The residual and grey waste fraction is further processed in a mechanical plant. The processing includes pre-shredding, screening, removal of non magnetic and magnetic metals, shredding, other magnetic removal and drum screening (50 mm). The fraction > 50 mm is processed to produce REF, while the sieved (< 50 mm) fraction was used in this study as the M residual. The M residual was transported in trucks to the landfill site operated by Mustankorkea Ltd (Jyväskylä, Finland) and was stored for two-three days outdoors before landfilling into lysimeters.

The MB residual was prepared by composting of M residual (described above) in seven batches for 2-3 weeks in aerated pilot tunnels (two 50 m^3 tunnels, Vapo Biotech Ltd, Jyväskylä) followed by pile composting and storing outdoors in piles for 6-14 months. The compost was mixed by a front loader weekly during the tunnel composting and two-three times in the first 2-3 months of the pile composting. Wood chips (0.5 m^3/t M residuals) or the composted oversize fraction (>15 mm, 0.5 m^3/t M residuals) of the MB residual was used as support material in the tunnel compost. The gas generation within 21 days of testing (GB21 value) was 22 Nl/kg TS after 3 weeks' tunnel and 5 weeks' pile composting as determined from one of the seven batches (Lehtinen 2003). This would have been close to the requirements presented in the German landfill ordinance (AbfAbIV 2001), which requires that mechanical biological treatment should stabilize waste so that its gas production potential over a period of 21 days measured by a standardised method (GB21) should be <20 Nl/kg TS. Finally, all the composted MB residuals were mixed together and screened in the drum (40 mm) to remove the support materials.

The temperatures within the landfilled M and MB residuals were monitored by a soil temperature and moisture station (Davis 6343) and temperature probes (Davis 6470) with a wireless Vantage Pro console (Davis 6310). Two parallel series (four probes per series) of temperature probes were located at two sites in both lysimeters (Fig. 3) and the results for each depth were reported as mean values of two parallel probes. Ambient air temperature was monitored by a weather station (Davis Vantage Pro 6150 equipped with a datalogger/PC-link 6510) on the landfill area. Rainfall data were obtained from the Finnish Meteorological Institute (2006).

3.1.4 Waste sampling and processing (I and V)

The sampling from landfill bodies was done during the installation of vertical gas collection wells (borehole Ø 1.2 m, Ämmässuo) or leachate observation tubes (borehole Ø 90 mm, Kujala) (I). The sampling procedures were planned to obtain depth profiles at different locations using a 50-100 m grid (Fig. 1-2). The volume of each sample was about 300–500 l in Ämmässuo and 10–20 l in Kujala. Samples were spread out on a concrete floor in layers of 20-40 cm from which two replicate randomised composite samples (about 30 l) were shoveled into plastic bags, one for sorting/description and the other for analyses in Ämmässuo, while in Kujala replicate samples were not taken. In Ämmässuo the sorting was done within two days, while for the analyses the samples were stored outside (below 0°C) for 1-2 months, where as in Kujala the samples were stored in airproof plastic bags outdoors (below 0°C) for 2-3 months before sorting and analyses. The drilling procedure during the construction of monitoring wells crushed most samples into particles of < 50 mm diameter in Kujala, while samples taken from Ämmässuo were less crushed during drilling. Some bulk materials (mainly stones, metals and glass) which are difficult to shred were removed and the proportion of these materials was included in the sorting results, but excluded from the analyses.

Waste samples were manually sorted into seven (Ämmässuo) or five (Kujala) categories, namely plastics, paper and cardboard, wood, metals, inert materials (e.g., glass, stones), textiles and residuals (I). Textiles were included in the residuals and metals in the inert materials when sorting the Kujala samples. Sorting was based on visual inspection, and thus small particles which could not be visually identified were classified as "residuals". Each category was weighed separately.

For analytical purposes the samples taken from Ämmässuo and Kujala were shredded (Retsch SM 2000) into three size fractions, except bulky materials which were not processed (e.g., metals and stones) (I). The particle size of the processed fractions was <2 mm for total Kjeldahl nitrogen (TKN) and <20 mm for the TS and VS analyses, and <50 mm for the methane production assays and shaking leaching tests. The bulky materials that were removed were weighed and taken account in the sorting, but not in the analyses. This means that compared to the actual landfill conditions some of the variables (e.g., TKN and BMP) in samples had higher observed concentrations relative to their in situ condition, according to the proportion of materials removed.

In the landfill lysimeter study (V) the M residual sample (60 l) was prepared by combining six 10 l samples obtained one from each 13-15 t truck container used for transporting the material to the test area, while the MB residual sample was combined from five 10 l samples obtained from randomly selected locations in the mixed MB residual pile formed when preparing the material for filling of the lysimeter (V). The characteristics of the M and MB residuals are shown in Table 5.

3.1.5 Leachate sampling (II and V)

The leachate was sampled in the monitoring wells and in the leachate (recirculation) well by bailers (Clear-View, high density polyethylene, volume 1 l) in Kujala landfill (II). Approximately 2-3 liters of leachate were drawn from the monitoring wells before the samples for the analyses were taken. In the landfill lysimeters (Mustankorkea landfill) leachate was sampled in the flow meter wells by a vessel (high density polyethylene, volume 2 l) connected to a rod (length 1.5 m).

3.2 Modelling methane generation (IV)

The methane generation rates for the Kujala and Ämmässuo landfills as a whole were modelled using the Landgem 3.02 landfill gas emission model (equation 1, USEPA 2005b). Besides annual landfilling rates (t waste /year) the default k and L_o constants provided with the Landgem model (k varying from 0.02 to 0.7 and L_o varying from 96 to 170 m^3/t depending on either values based on inventories or determined in the Clean Air Act (CAA) and landfill location in wet or arid areas

were used as well as k and L_o determined experimentally on the basis of the BMPs (used as L_o in model) determined in batch assays (I). The L_o was approximately 110 and 30 m^3/t (wet weight) in the top layers in Ämmässuo and Kujala. In addition to these values, methane generation was modelled using higher L_os (130, 150 and 170 m^3/t for Ämmässuo and 40, 50 and 60 m^3/t for Kujala) on the grounds that the contemporary L_os were probably higher than those in the 2- to 3-year-old samples used in batch assays. The ks were determined on the basis of the half-life time ($t_{1/2}$) of L_o assuming that it corresponds to the mass-based half-life time of degradable organic carbon and using the previously reported (IPCC 2006) relationship between k and $t_{1/2}$ for degradable organic carbon: $k=\ln(2)/t_{1/2}$. For Ämmässuo half-life was calculated using the linear trend between the L_o of waste landfilled in 2003 (average approximately 110 m^3/t, in the upper layer of landfill sector 1) and L_o of waste landfilled around 1987 (average approximately 10 m^3/t in the bottom layer of sector 3). For Kujala the half-life of L_o was calculated on the basis of the L_o of waste landfilled in 1998-2003 (average 30 m^3/t in the upper layers) and L_o of waste landfilled in 1984-1989 (average 15 m^3/t in one of the bottom layers) located approximately 7 to 9 meters from the bottom structure. Moreover a k similar (0.05, 0.06, 0.07 for Ämmässuo and 0.03, 0.04, 0.06 for Kujala) or close to the k of 0.04 previously determined for conventional landfills (USEPA 2005b) was screened in the present study for both landfills with previously determined L_os of 110 and 30 m^3/t.

$$Q_{CH4} = \sum_{i=1}^{n} \sum_{j=0,1}^{1} kL_o \left(\frac{M_i}{10} \right) e^{-kt_{ij}} \qquad (1)$$

Where:
Q_{CH4} = annual methane generation in the year of calculation (m^3/a), i = 1 year time increment, n = (year of calculation) - (initial year of waste acceptance), j = 0.1 year time increment, k = methane generation rate ($year^{-1}$), L_o = methane generation potential (m^3/t), M_i = mass of waste accepted in the i^{th} year (t) and t_{ij} = age of the j^{th} section of waste mass M_i accepted in the i^{th} year (decimal years).

3.3 Analyses, determinations and measurements (I, II, V)

BMPs were determined with digested mesophilic municipal sewage sludge (Nenäinniemi Jyväskylä, Finland) as an inoculum (500-1300 ml) in duplicate 2 l glass vessels (I, V). The waste samples were added to the vessels to obtain the ratio of 2 g VS_{waste}/g $VS_{inoculum}$, at a ratio which usually enables methane generation without the accumulation of organic acids. The BMP of the inoculum was determined separately and was reduced from that of the samples and inoculum together. The final liquid volume of 1.5 l in each vessel was obtained by adding deionised water; thus the moistures in all the assays (with waste, inocu-

lum, and water) were 88 - 95 % of wet weight. The bottles were flushed with N_2/CO_2 (80/20 %) and sealed with butyl rubber stoppers. The gas produced was collected in aluminium sampling bags (Tecobag PETP/AL/PE-12/12-75, Tesseraux Spezialverpackungen). The BMPs were continued until methane production became neglible (<5 ml CH_4 d^{-1}) after 70-100 d.

TS and VS were analysed according to standard methods (APHA 1988). Moisture content was counted by subtracting the analysed TS content from 100 %. Leaching of NH_4-N and COD were determined by samples obtained in a one-stage shaking leaching tests at a liquid-to-solid (L/S) ratio of 10 according to SFS-EN 12457-4 (2002), except that the samples were shredded below 50 mm, instead of the 10 mm particle size mentioned in the standard method. The samples for the NH_4-N and COD analyses were filtered with GF50 glass fibre filter papers (Schleicher & Schuell). COD was analysed according to SFS 5504 (1988). TKN and NH_4-N were analysed according to the application note supplied by Tecator (Perstop Analytical/Tecator AB 1995). A WPA CD70 pH meter and Sensorex pH 450 CD electrode were used to measure the pH of the Ämmässuo samples. Biogas volume produced in methane potential assay was measured using a water displacement method in a special gas measurement cylinder. Methane content was measured by a Perkin Elmer Autosystem XL gas chromatograph with a flame-ionization detector as described in Luostarinen & Rintala (2005) in the methane potential assays. Pore gases (CH_4, CO_2 and O_2) were measured by an IR analysator (Geotechnical Instruments GA 94) with steel tubes (Ø 30 mm) at depths of 0.5, 1.0 and 1.5 m in the landfill lysimeters. Gas emissions (CH_4, CO_2, N_2O) were measured by a FTIR analysator (Gasmet DX4000) at five sampling points (distance about 2 m) with a flow chamber (Ø 50 cm and volume 29 l) method in mechanically and mechanically-biologically treated municipal solid waste lysimeters.

4 RESULTS

4.1 Characterisation of landfills by waste sampling (I)

4.1.1 Waste composition

The two landfills, Ämmässuo and Kujala, which had been in operation for approximately 20 and 50 years, respectively, were sampled in order to determine vertical and horizontal variability in structure, composition and properties. Clear trends appeared mainly with respect to sampling depth, although some differences between individual samples were also observed horizontally (Fig. 4-6). However no clear spatial differences were detected (data not shown) horizontally across the landfills.

In Ämmässuo the major weight fraction was inert materials (30-40 %, mainly stones), while the other fractions contributed shares of <22 % (Fig. 4). In Kujala the residuals (soils and unrecognisable materials) formed the largest fraction at all depths (54-75 %). The proportion of paper and cardboard was lowest in the bottom layer of both landfills (2 % in Ämmässuo and 0.5 % in Kujala) while the percentage of wood (15-16 % in Ämmässuo and 9-13 % in Kujala) was almost the same at all depths in both landfills.

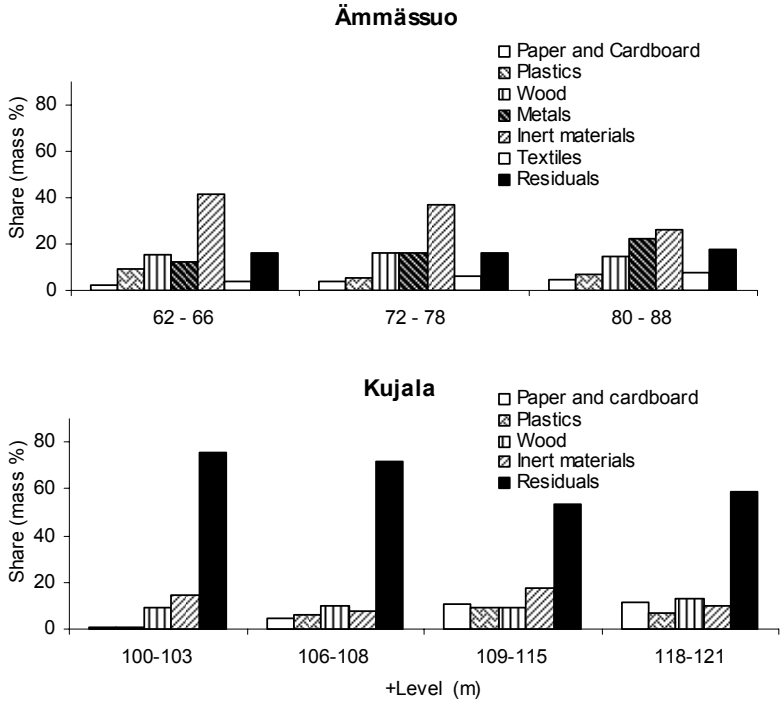

FIGURE 4 Waste composition at different depths in Ämmässuo and Kujala landfills (I).

4.1.2 Totals solids, organic matter and nitrogen contents

The TS, VS, VS/TS ratio, pH (measured from the Ämmässuo samples) and TKN of the landfill body samples were studied to evaluate the properties and conditions within the landfills (Fig. 5). In Ämmässuo pH varied from 6.7 to 8.9 and the mean pH at different layers increased from 7 to 7.6 towards the bottom of the landfill. In Kujala the pH of the leachate (measured from observation tubes in the same location from which the samples were obtained) ranged from 6.8 to 8.2. The TS of individual samples ranged from 35 to 81 % in Ämmässuo and from 56 to 89 % in Kujala, while mean TS content was lower in Ämmässuo (54 %) than Kujala (68 %). Mean TS decreased (from top down) in Ämmässuo while in Kujala no clear trend along the depth profile emerged. The mean VS/TS ratio of samples was 63 % in Ämmässuo (range 35 to 89 %) and 40 % in Kujala (range 3 to 80 %). In both landfills the VS/TS ratio was lowest (mean 55 % in Ämmässuo and 16 % in Kujala) in the bottom layers. The TKN of individual samples ranged from 1.3 to 7.8 g/kg dry weight (mean 3.9 g/kg dry weight and 2.1 g/kg wet weight) and from 0.8 to 9.3 g/kg dry weight (mean 4 g/kg dry weight and 2.7 g/kg wet weight) in Ämmässuo and Kujala, respectively. The mean TKN of the bottom layers were 52 and 60 % (% dry weight) that of the top layers in Ämmässuo and Kujala, respectively.

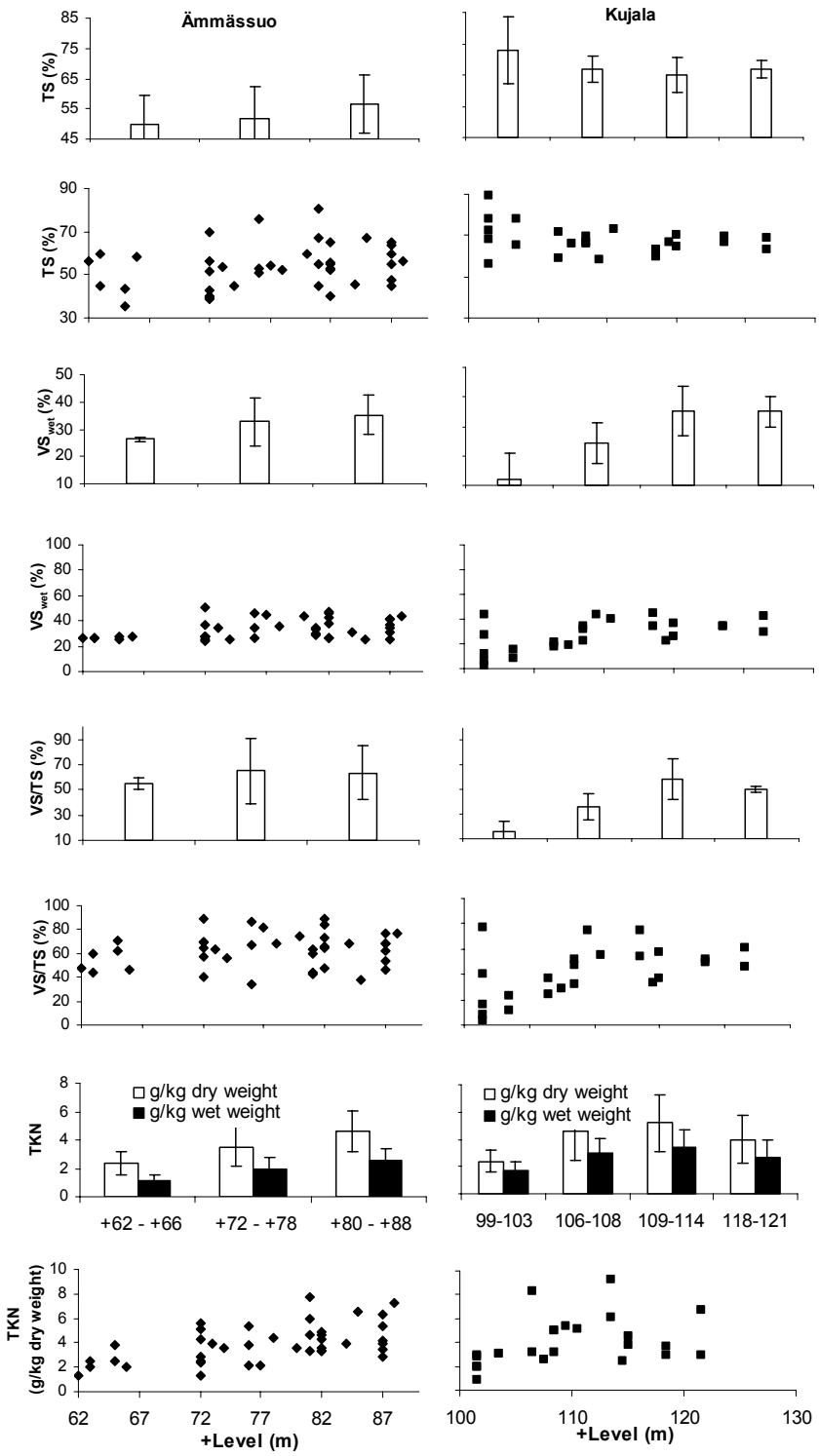

FIGURE 5 TS, VS, VS/TS and TKN contents of waste samples along the depth profiles and mean (± stdev) values at different layers of Ämmässuo and Kujala landfills (I). The levels measured from the mean sea level.

4.1.3 Leaching of nitrogen and organic material and methane production properties

The leaching of NH_4-N and COD in the landfill body samples were studied by shaking leaching tests. NH_4-N-leaching was 0-2205 g/t TS (mean 841 g/t TS) in Ämmässuo and 0-1400 g/t TS (mean 390 g/t TS) in Kujala (Fig. 6). In Ämmässuo the highest NH_4-N leaching was in the middle layer of the landfill and in Kujala the highest NH_4-N leaching was in the second (from top) layer. The NH_4-N-leaching/TKN ratio was 0-79 % (mean 25 %) in Ämmässuo and 0-24 % (mean 9 %) in Kujala. The highest NH_4-N leaching/TKN ratios were in the middle and bottom layer in Ämmässuo and in the two middle layers in Kujala, in spite of the fact that nitrogen content was lowest in the bottom layers of both landfills. COD leaching was about 4-fold higher in Ämmässuo (2.6-51.4 kg/t TS, mean 19.3 kg/t TS) than in Kujala (0.5-15.9 kg/t TS, mean 5.6 kg/t TS). In Ämmässuo COD leaching decreased downwards in the landfill, while in Kujala the highest COD leaching was in the second (from top) layer.

The BMPs of the waste samples and the effect of water addition on the methane production rate were determined in batch assays. The BMPs ranged from 1 to 183 m^3/t TS (mean 50 m^3/t TS) in Ämmässuo and 6-60 m^3/t TS (mean 34 m^3/t TS) in Kujala (Fig. 6). In both landfills BMP was lowest in the bottom layer, while wide variation and the highest values (above 100 m^3/t TS in Ämmässuo and above 40 m^3/t TS in Kujala) were found in the upper layers. The methane production of four samples from both landfills at their original moisture (33-57 % in Ämmässuo and 32-37 % in Kujala) content and at 60% moisture content started after a lag of 2-3 months (Fig. 7). Methane production at original moisture content was 0.0006-8.3 m^3/t TS (Ämmässuo) and 0.03-0.2 m^3/t TS (Kujala) after incubation for 544 (Ämmässuo) and 372 (Kujala) days. The adjustment of moisture content to 60 % increased methane production to 1.6-63 m^3/t TS (Ämmässuo) and 0.06-13.4 m^3/t TS (Kujala), the highest increase for an individual sample being from 8.3 to 63 m^3/t TS for a sample with 50 % original moisture. In the other words, methane production at the original moisture content was 0.0003-4.5 % (Ämmässuo) and 0.2-1.0 % (Kujala), while at 60 % moisture content methane production was 2.3-34 % and 0.9-32 % of BMP in Ämmässuo and Kujala, respectively.

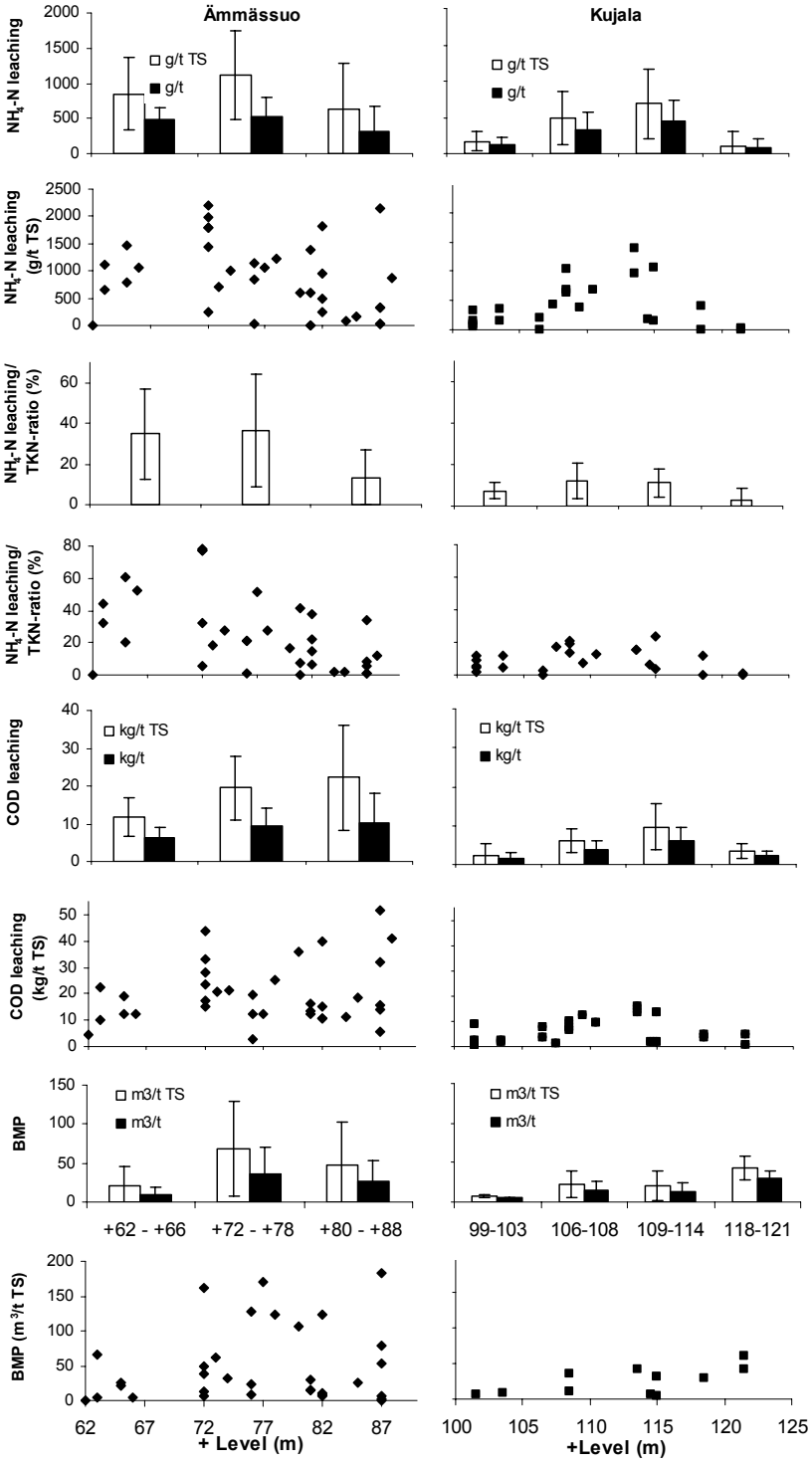

FIGURE 6 NH_4-N leaching, NH_4-N leaching/TKN ratio, COD leaching and BMP of waste samples along the depth profile and mean values (± stdev) at different layers of Ämmässuo and Kujala landfills (I). The levels measured from the mean sea level.

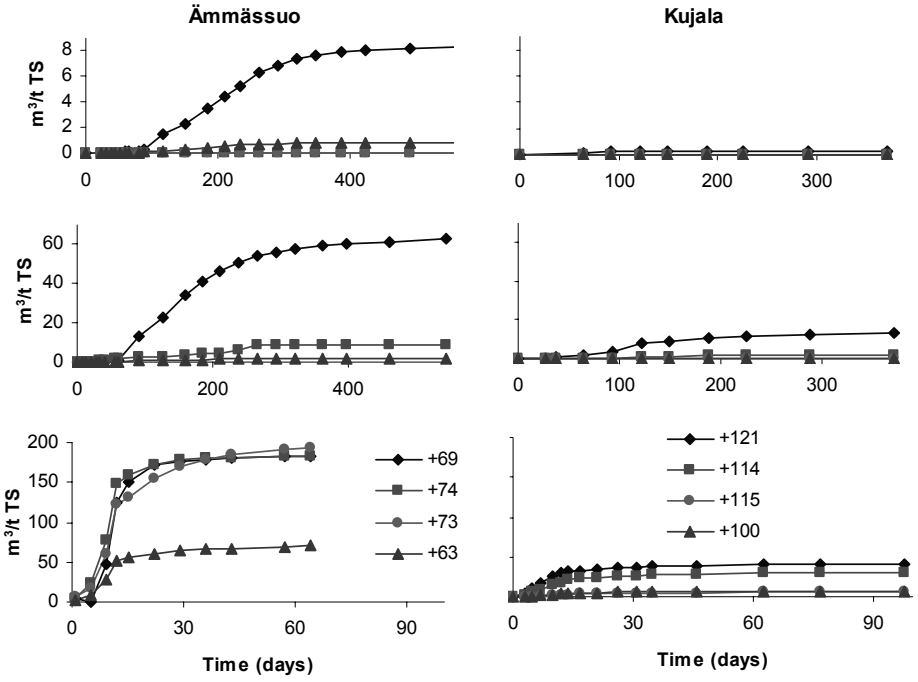

FIGURE 7 Methane production from Ämmässuo and Kujala samples (named in figure by sampling depth above mean sea level) at original moisture content (up), at 60 % moisture (without additional inoculum; middle) and in BMP assay (inoculum added; bottom) (I).

4.2 Characterisation of internal leachate quality in a municipal solid waste landfill (II, III)

4.2.1 Leachate quality and its vertical profile (II, III)

For this study 14 monitoring wells were installed in Kujala landfill over an area of approximately 2 ha in order to characterise leachate quality within the MSW landfill and its vertical, horizontal and temporal variation as well as impacts of lechate recirculation during a period of approximately 2 years (II). In addition to manual sampling and characterisation by analyses four monitoring wells in each landfill were furnished with an on-line monitoring system in order to characterize changes in the water table and temperature (III). In addition, in Kujala, the feasibility of on-line measurements of electrical conductivity and pH was studied.

Table 6, which summarises (mean ± std) the leachate characteristics in the 12 different monitoring wells (Fig. 2) and in the leachate recirculation well during the 2-year study period, shows that the mean concentrations and values of the studied parameters varied between different wells and also that variation occurred within most of the wells during the study. For example, the lowest and highest individual COD values measured were about 400 mg/l and 10 000

mg/l, while the lowest and highest mean values in different wells were 642±46 mg/l (E1) and 8037±1138 mg/l (E2). In general, the differences between different wells were much higher (as percentages) in COD and BOD than in the other parameters (e.g., NH_4-N, Cl).

The water table (measured from mean sea level) varied between the monitoring wells (from +102.6 to 103.7 in H1; from +103.8 to +103.9 in C1; from +108.7 to 109.0 in C3; from +110.0 to +110.7 in G2 and from +104.0 to 104.9 in G3). This probably had an effect on leachate quality via sampling depth as the mean zone of saturation varied from approximately 20 % (in C1) to 51 % in (G2) of the height of the landfill, which meant that the screens of the low level monitoring wells (C1, E1, G1; with short screen 2 m) were in the saturated part while the screens of the higher level monitoring wells (with long screen from 6.5 to 21 m) were partly in the non-saturated part.

The leachate characteristics were studied as a function of the level of the monitoring well (assumed to gather water from low or high levels of the landfill; Fig. 8). The mean concentrations or values of most parameters in the leachate recirculation area were lower in the two low level monitoring wells (E1, G1) than at higher levels, although one low level well (C1) also had higher values. On the other hand, low values were observed also in high level wells in the reference area (H1, H2). The highest NH_4-N concentrations and COD as well as BOD values were observed in well E2, which, however, had a well profile comparable to that of most of the wells (e.g., B1, D1, F1 and G3). Moreover E2 also had significantly higher values compared to E1, located only 5 m away from it but gathering leachate from the lower level of the landfill (E1 99.5-101.5 m, E2 104.94-122.94 m). The BOD/COD (means from 0.08 to 0.17 in different monitoring wells) and NH_4-N/TKN (means from 0.75 to 0.90 in different monitoring wells) ratios as well as pH (from 6.9 to 8.5) did not vary significantly with landfill level.

TABLE 6 Landfill height at locations of monitoring wells, screen range of monitoring wells and characterisation (mean±stdev, except pH range) of leachate samples (II).

Well	Height [1] (m)	Screen range (m)	Distance from recirculation canals (m)	Conductivity (mS/m)	pH (range)	NH_4-N (mg/l)	TKN (mg/l)	COD (mg/l)	BOD (mg/l)	Cl (mg/l)	BOD/COD ratio
C1	24.6	99.9-101.9	5	1963±317	7.4-8.1	1536±501	1863±497	4651±1681	338±182	1503±226	0.11±0.03
C2	25.1	104.1-122.1	5	na[2]	7.3-7.7	1352±275	1968±668	na	na	1357±75	na
D1	25.3	103.3-122.3	5	2080	7.8-8.1	1765±288	2260±498	4269±1348	774±230	1417±248	na
F1	22.5	103.1-119.1	5	2235±279	na	1809±91	2207±133	6422±389	748±45	1561±49	0.11±0.007
G1	21.5	98.7-100.7	5	716±88	6.9-7.2	505±294	479±79	1030±862	53±14	436±102	0.08±0.03
G3	19.6	101.5-115.2	5	1819±193	7.3-7.8	1311±503	1457±448	4152±909	580±319	1427±271	0.16±0.11
E1	24.4	99.5-101.5	25	1282±891	7.9-8.1	271±31	286±85	642±46	75±10	475±53	0.11±0.01
E2	24.6	104.9-122.9	25	2206±256	7.7-8.5	2099±209	2553±308	8037±1138	1301±483	1671±125	0.17±0.07
B1	24.8	101.9-119.9	25	2139±165	7.6-7.9	1676±391	2082±520	5172±931	614±114	1636±169	0.11±0.02
A1	22.4	102.4-119.4	45	2113±102	na	1835±211	2329±536	4427±489	494±106	1442±242	0.11±0.04
H1	13.2	99.9-109.9	55	1468±143	7.4-7.7	757±132	881±134	1446±245	106±33	1113±188	0.09±0.05
H2	8.6	100.8-107.3	60	na	na	464-130	427±82	822±136	105±24	567±34	0.12±0.05
LW[3]				402±122	7.2-8.7	273±540	131±54	363±121	1310±66	344±230	0.13±0.11

[1] Height of the landfill body; [2] na =not analysed; [3] LW (leachate recirculation well)

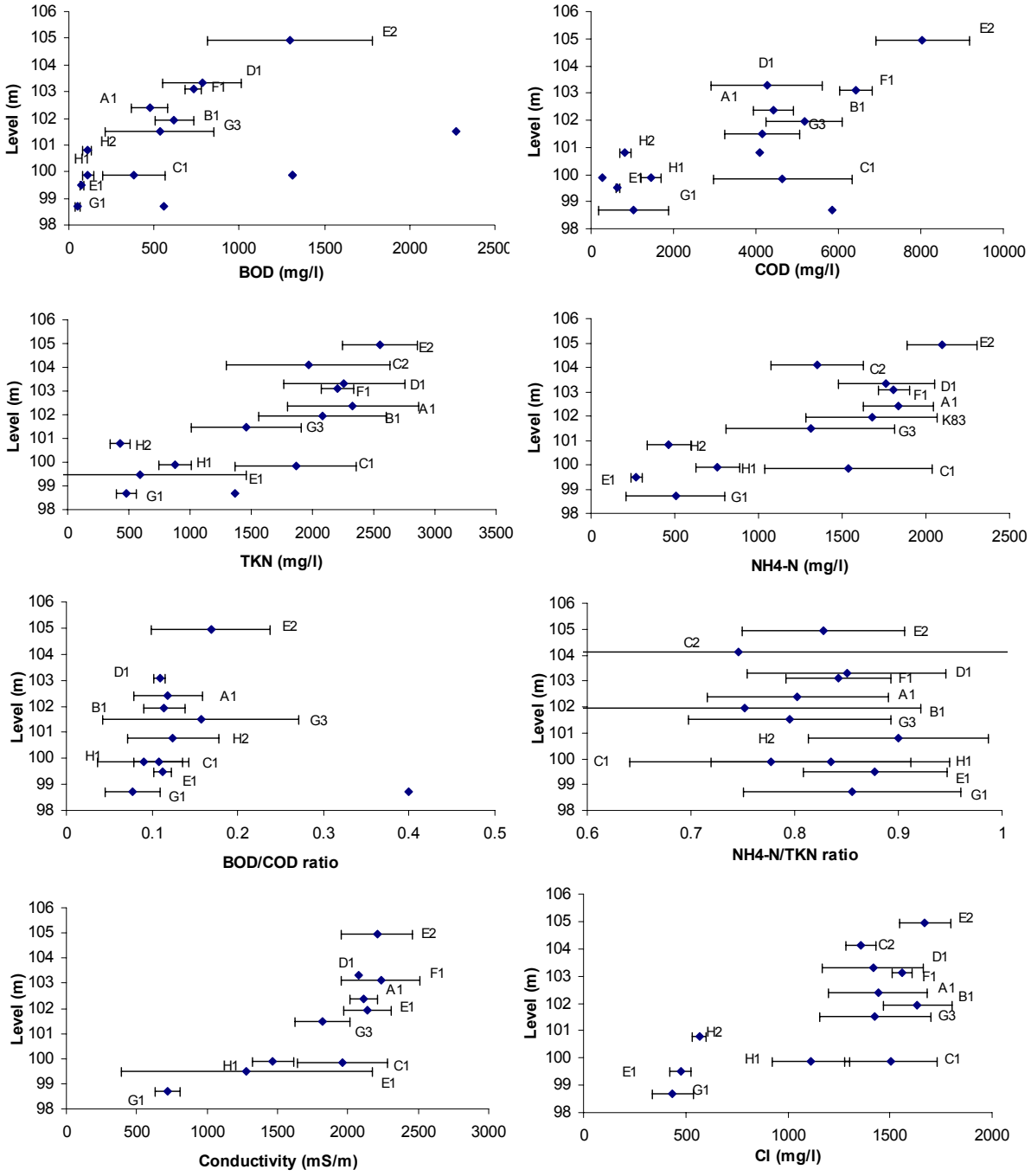

FIGURE 8 Mean concentrations of BOD, COD, TKN and NH_4-N, BOD/COD and NH_4-N/TKN ratios as well as mean conductivity and chloride concentrations in leachate obtained from monitoring wells located at different levels in the landfill body (II).

4.2.2 Temporal and horizontal variation in leachate quality (II, III)

Temporal variation in leachate quality during the study period was also analysed in order to not only evaluate temporal changes and trends in leachate quality but also the potential impacts of leachate recirculation on leachate quality (Fig. 9-10). The concentrations and values of different parameters in the monitoring wells at varying distances (from 5 to 25 meters) from the leachate recirculation canals varied randomly and were not dependent on the distance of the monitoring well to the recirculation canal. However, the variation in the COD/chloride ratio in the two low level monitoring wells (C1 and G1) and in the higher level well (G3), located at 5 meters distance from the leachate recirculation canal might indicate that the recirculated leachate was diluting the chloride content in the leachate in the landfill body, as the concentrations of chloride in the recirculated leachate, which was collected from the whole landfill area, were lower (252-500 mg/l, mean 345 mg/l) than in the leachate in the landfill body (300-2100 mg/l, mean 1217 mg/l). However, the leachate quality in G1 varied less than in monitoring wells C1 and G3 on the basis of the electrical conductivity, which may also indicate local effects of leachate recirculation on leachate quality in certain monitoring wells.

4.2.3 On-line monitoring of internal leachate (III)

Variation and seasonal changes in the water table and temperature were studied by on-line measurements in four monitoring wells (A, B, C, D) in Ämmässuo landfill for periods between 194 and 609 days (Fig. 11). The highest and lowest measured water table and temperature in the four monitoring wells ranged from 3.5 to 9.5 m and from 10 to 39 °C, respectively. The water table varied within two meters in same monitoring wells during the study. The water table increased gradually in B, C and D, while in A the water table fell by 1.5 m during two months (April and May) in 2003 and varied thereafter within 0.5 m during the next ca 14 months. The highest variation in temperature of 12 °C (from 39 to 27 °C) was observed with a decreasing trend in A, while in B the temperature varied within 8 °C (from 12 to 20 °C) without showing a clear trend and in C and D temperature varied within 2 °C, in the latter with an increasing trend.

Water table and temperature were measured (on-line) in four (C3, G2, G3 and H1; 212-516 days) conductivity in three (C1, G3 and H1; 167 days) and pH in one monitoring well (H1; 167 days) in Kujala. The water table varied within approximately 8 meters between the four monitoring wells, while in the same monitoring wells the variation was mainly within one meter; for example, the water table increased slowly from 104 m to 105 m in G3 and H1. Temperature varied from 17 to 35 °C between the four monitoring wells and in same monitoring well from 2 to 10 °C.

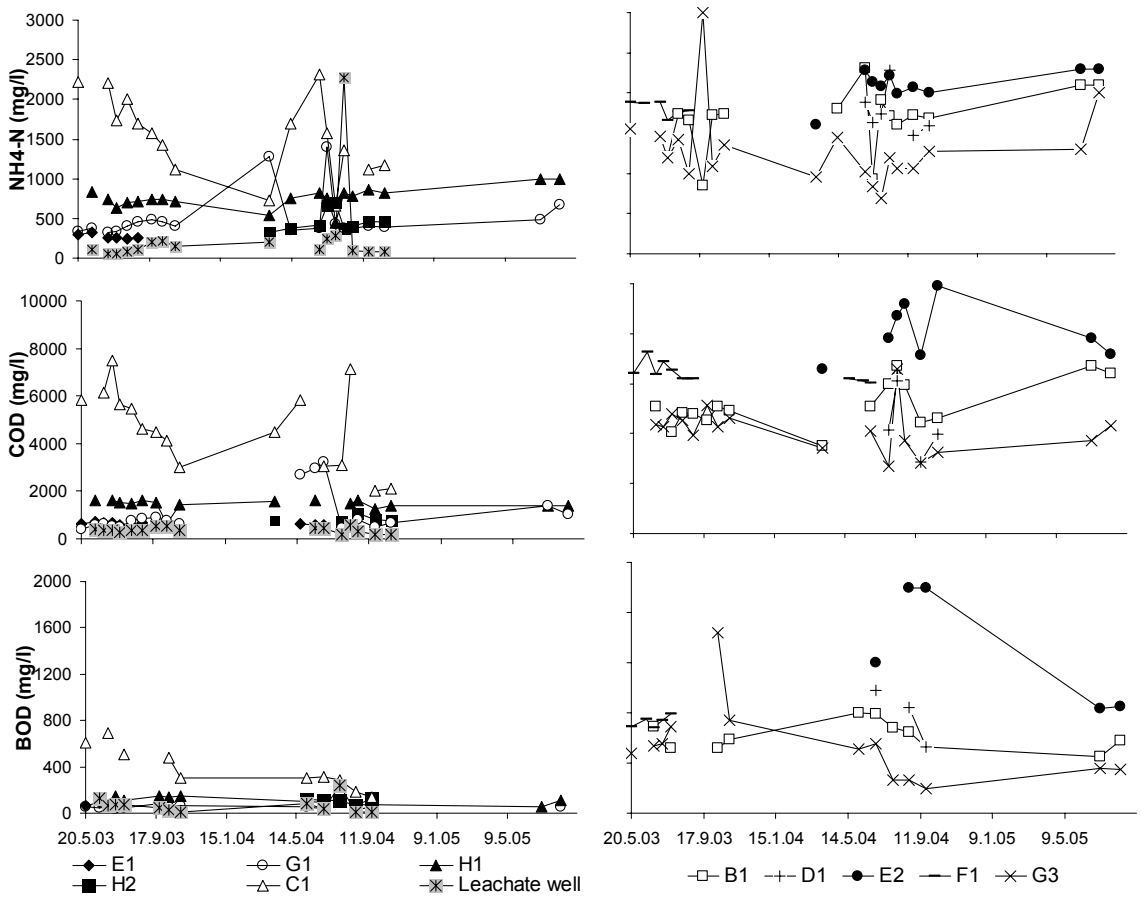

FIGURE 9 NH_4-N, COD and BOD in low (left) and high (right) level monitoring wells and in leachate (recirculation) well (II).

The highest temperature was recorded in C3 (range 33 - 34 °C), while the highest individual value (35 °C) was measured in G3, where the temperature however decreased gradually during the study to 29 °C. In other two monitoring wells the temperature was lower, increasing from 23 to 26 °C in G2, and decreasing from 19 to 17 °C in H1.

Electrical conductivity was measured (on-line) in three (C1, H1, G3) monitoring wells and pH in a monitoring well (H1). The average conductivity was higher and varied more in G3 (1300-2300 mS/m) than in the other monitoring wells studied (1300-2000 mS/m, C1; 1300-1600 mS/m, H1; Fig. 12, 14) for five months. The variation in G3 coincided with the recirculation of more dilute leachate (conductivity of 300-700 mS/m) in the recirculation canal located at five meters from G3. Only minor variation was observed in pH, which ranged from 6.9 to 7.3, without showing a clear trend, in H1 (Fig. 13).

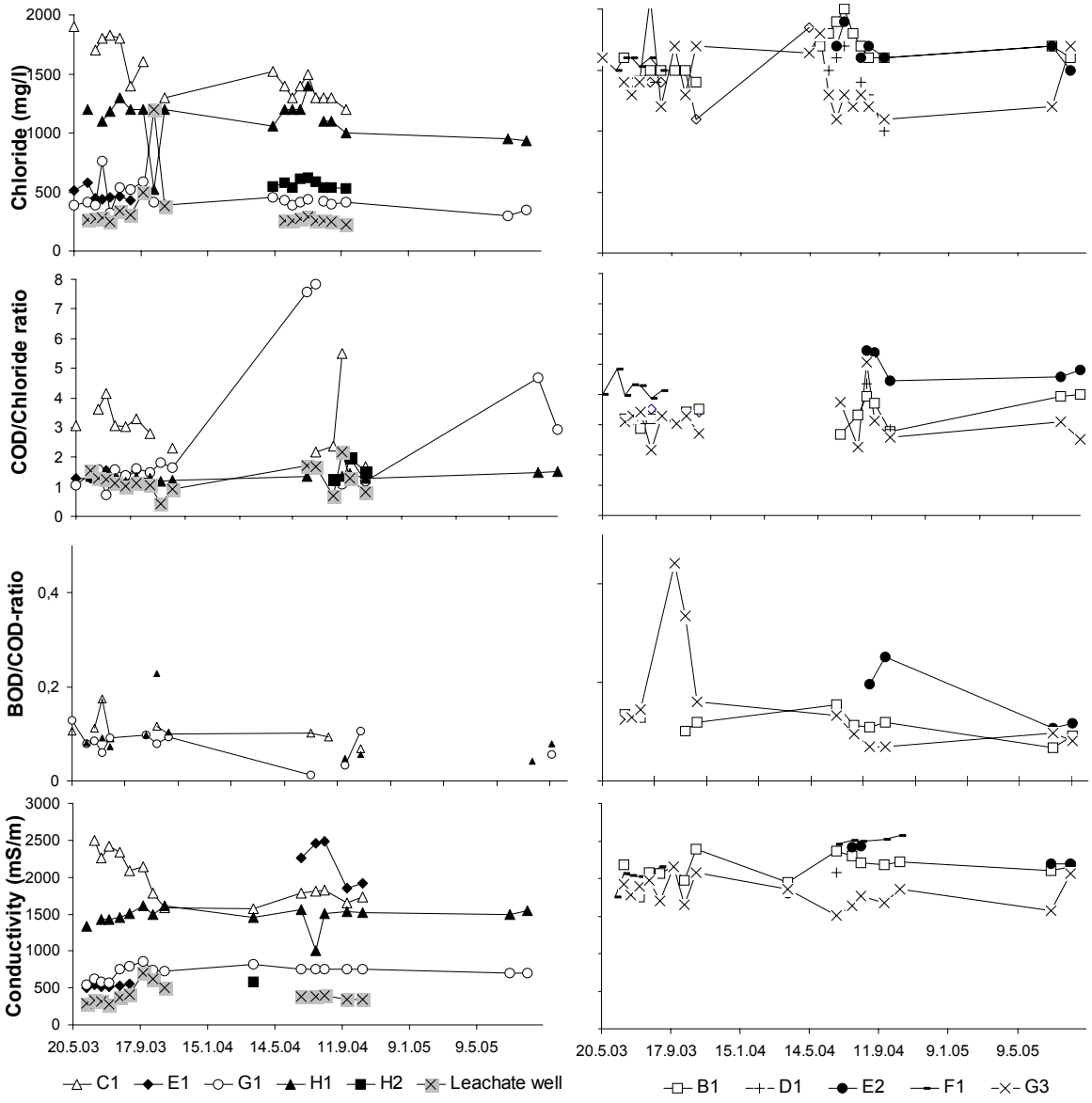

FIGURE 10 Chloride, COD/Chloride and BOD/COD ratios and electrical conductivity in low (left) and high (right) level monitoring wells and in leachate (recirculation) well (II).

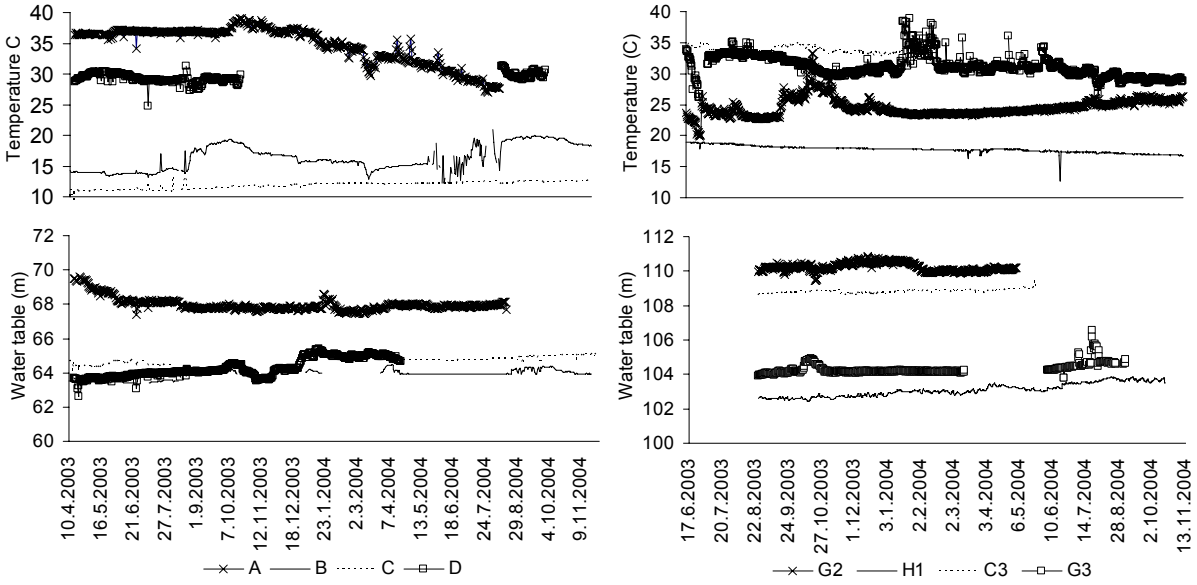

FIGURE 11 On-line measurements of temperature and water table at monitoring wells in Ämmässuo (left) and Kujala (right) landfills (III).

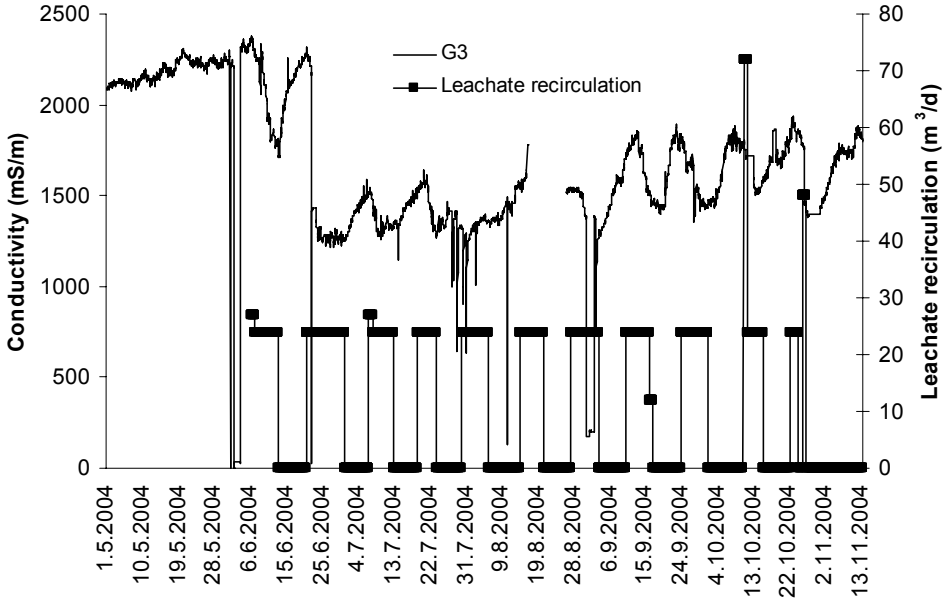

FIGURE 12 Electrical conductivity in leachate in the monitoring well (G3) during the period of leachate recirculation in 2004 (III).

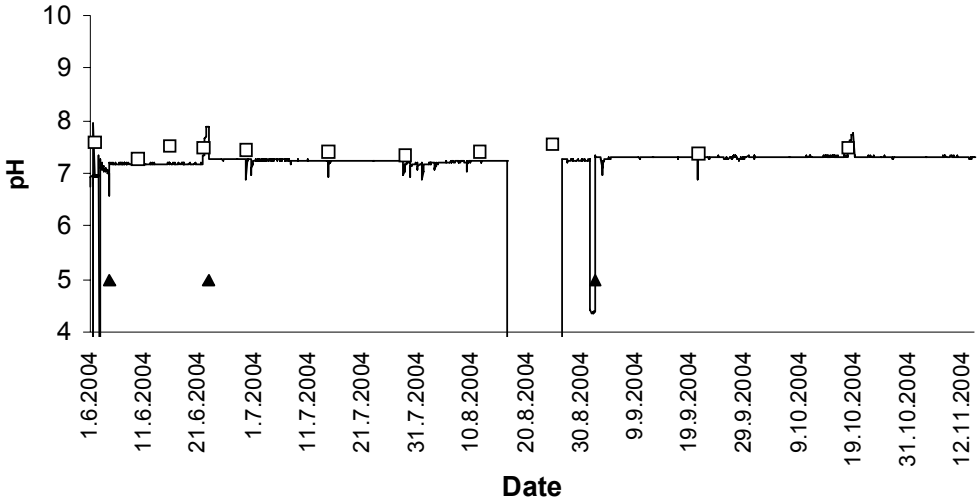

FIGURE 13 On-line (--) pH, calibrations (▲) and pH of leachate samples measured by field meter (□) at observation tube H1 (III).

4.2.4 Comparison of on-line and manual measurements (III)

The feasibility of monitoring the pH and electrical conductivity of leachate by on-line measurements was studied in instrumented monitoring wells in Kujala landfill. The on-line pH values (H1) were consistently 0.2 to 0.3 higher during the 5-month period than the manual values, while both measurements showed similar trends (Fig. 13). During 31.8-1.9.2004 on-line pH showed values between 4.3 and 4.4 due to fact that the electrode was in buffer solution (pH 4) outside of the monitoring well owing to testing for a sudden change in pH and for calibration. On-line and manual measurements of electrical conductivity were compared over a period of 5 months in three monitoring wells (Fig. 14; C1, H1, G3). During the study the on-line measurements differed by less than 100 mS/m in C1 (on-line from 1340 to 1970 mS/m; manual from 1660 to 1840, excluding the first three weeks) and, mostly, by less than 200 mS/m in G3 (on line from 1220 to 2330; manual 1220 to 2860 mS/m) and less than 50 mS/m in H1 (on-line from 1260 to 1550, manual from 1000 to 1560, excluding the first three weeks) until the third calibration (1.9.2004) caused a systematic decrease of approximately 200 mS/m in the on-line level of conductivity compared to the values obtained by the field meter.

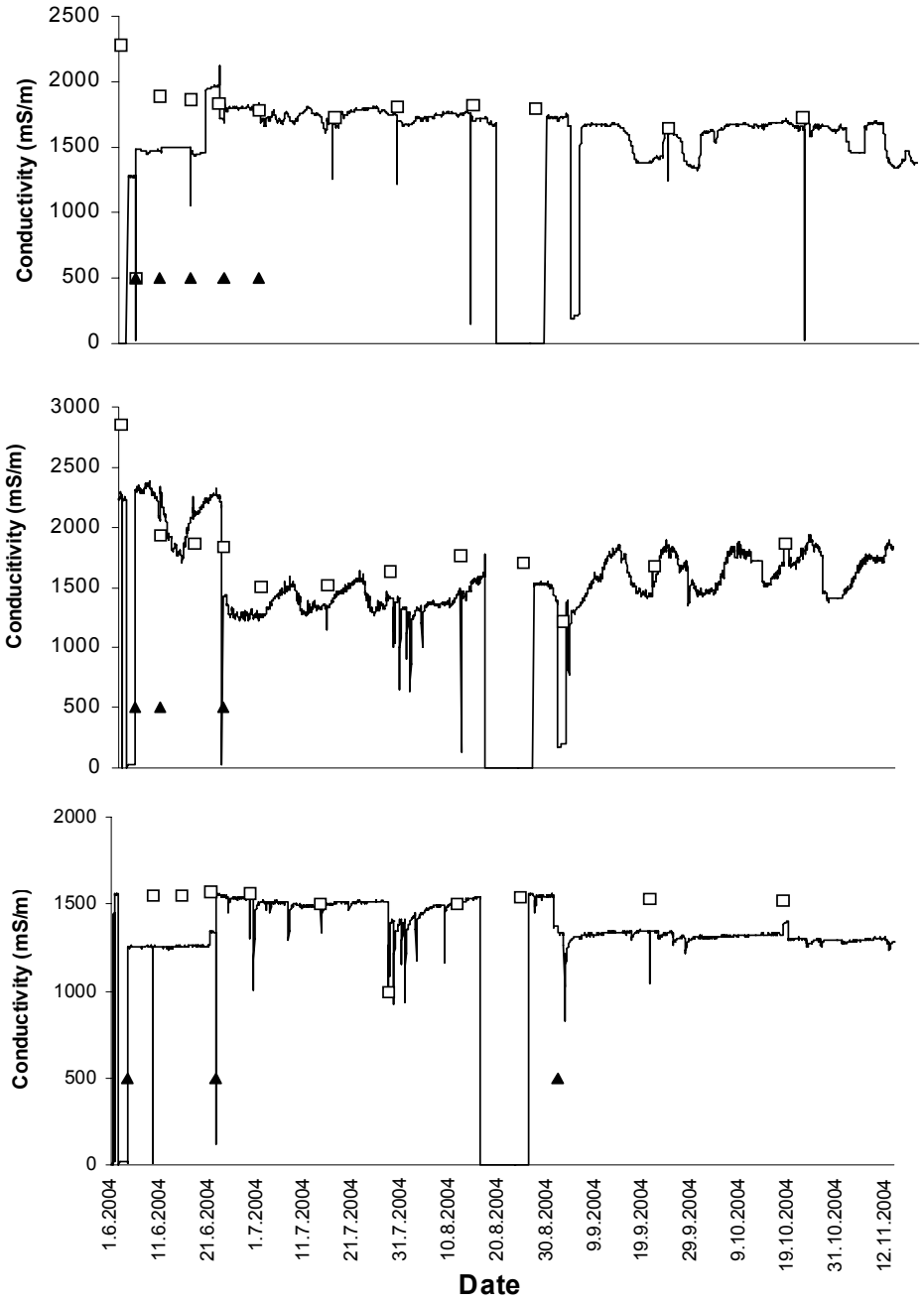

FIGURE 14 On-line electrical conductivity (-) in C1 (up), G3 (middle) and H1 (down), and calibrations of electodes (▲) and electrical conductivity of leachate samples measured by field meter (□) (III).

4.3 Determination of first order kinetic methane generation factors and methane generation potentials (IV)

4.3.1 Methane generation rate

First order kinetic methane generation factors (rate k and potential $L_\circ$ for bulk waste) were estimated for Ämmässuo and Kujala MSW landfills. In recent years, while both landfills have mainly been receiving source segregated residual fraction of MSW, their waste management and landfilling histories have been markedly different. Past and expected methane generation rates were modelled using the experimental data (I) obtained in the waste characterisation study and the methane recovery data obtained from the landfill operators.

The methane generation rate (k) for the two landfills was determined on the basis of the methane generation potential ($L_\circ$) and its half-life time ($t_{1/2}$) in studied landfills. In Ämmässuo $t_{1/2}$ of $L_\circ$ would be approximately four years and thus k 0.17 year^{-1}, while in Kujala $t_{1/2}$ of $L_\circ$ would be approximately 14 years and k 0.05 year^{-1} (Table 7).

TABLE 7 Determination of k on the basis of L_o and their half lives in Ämmässuo and Kujala landfills (IV).

Ämmässuo						
Years of landfilling	n	Mean TS (%)	Mean VS/TS (%)	Mean $L_\circ$ (m^3/t)[1]	Half-life of $L_\circ$ (years)	k (k=ln(2)/$t_{1/2}$)
2001-2002 (sector 1, top layer)	3	61	71	113	4	0.17
1987-1992 (sector 3, bottom layer)	6	50	55	11		
Kujala						
1998-2003 (top layer)	3	67	51	30	14	0.05
1984-1989 (7-9 m from bottom structure)	2	67	36	15		

[1]wet weight

4.3.2 Methane generation and recovery rates by varying methane generation factors

Methane generation in the studied landfills was modeled using differing values for k and the results compared to the actual amounts of methane recovered in those landfills. Modeling was performed using the experimentally obtained k of

0.17 for Ämmässuo and 0.05 Kujala as well as the values of 0.05, 0.06, 0.07 for Ämmässuo and 0.03, 0.04, 0.06 for Kujala with experimentally obtained L_o of 110 m^3/t for Ämmässuo and 30 m^3/t for Kujala (Table 8). The present methane generation varied from 31 to 50*10^6 m^3/year in 2006 and from 1.7 to 2.2*10^6 m^3/year in 2005 in Ämmässuo and Kujala, respectively, depending on the k used (Fig. 15). From 2007 onwards (both landfills will be closed in 2007) the methane generation seems likely to decrease faster rate in Ämmässuo than Kujala due to higher k; e.g., with k of 0.17 methane production from 2007 to 2008 will fall by approximately 9*10^6 m^3/a in Ämmässuo.

TABLE 8 The methane recovery rates (% of modeled methane production) as modelled by the Landgem (3.02) model with different k and L_o in Kujala and Ämmässuo landfills (IV).

Ämmässuo	L_o = 110 m^3/t, varying k from 0.05 to 0.17				k = 0.17, varying L_o from 130 to 170 m^3/t		
Year	L_o = 110 k = 0.05	L_o = 110 k = 0.06	L_o = 110 k =0.07	L_o = 110 k = 0.17	L_o = 130 k = 0.17	L_o = 150 k = 0.17	L_o = 170 k = 0.17
1996	20	17	16	9	8	7	6
1997	19	16	15	9	8	7	6
1998	16	14	13	8	7	6	5
1999	36	32	29	19	16	14	12
2000	61	54	49	32	27	24	21
2001	69	61	56	37	31	27	24
2002	82	72	66	44	37	32	28
2003	88	78	71	48	41	36	31
2004	99	88	80	56	48	41	36
2005	110	99	91	65	55	48	42
2006	122	110	101	75	63	55	48
Kujala	L_o = 30 m^3/t, varying k from 0.03 to 0.06				k=0.05, varying L_o from 40 to 60 m^3/t		
Year	L_o = 30 k = 0.03	L_o = 30 k = 0.04	L_o = 30 k = 0.05	L_o = 30 k = 0.06	L_o = 40 k = 0.05	L_o = 50 k = 0.05	L_o = 60 k = 0.05
2002	79	69	64	61	50	38	36
2003	88	77	71	68	54	43	41
2004	99	87	81	78	61	49	47
2005	102	91	85	81	63	51	49

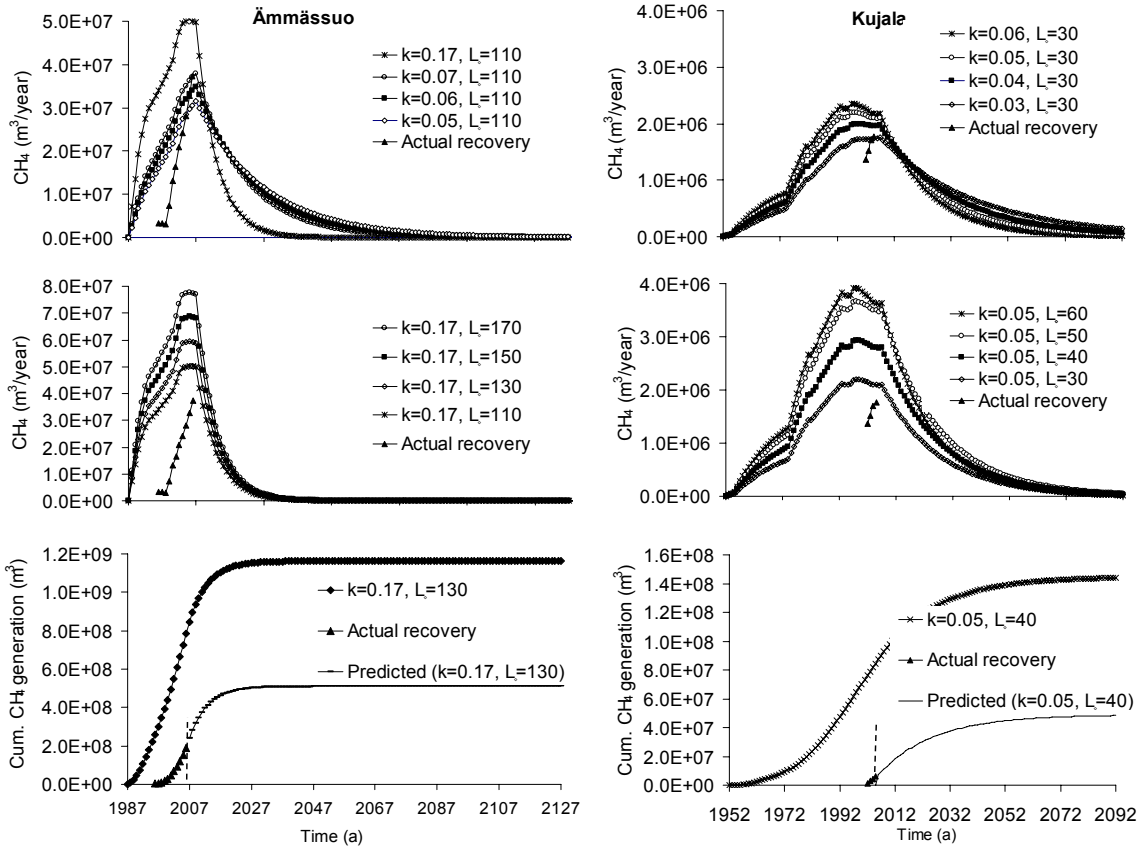

FIGURE 15 Methane production by varying k (top) and varying L_o (middle) compared to actual methane recovery and the modelled cumulative methane generation (bottom) b experimentally determined k and L_o in Ämmässuo and Kujala (IV).

The reliabilities of k and L_o for the two landfills were estimated by the proportion of methane recovery calculated according to the methane generation modelled with the varying k and L_o values. However methane recovery was increasingly implemented and optimized by gas characteristics (CH_4 and O_2 concentrations), gas flow and suction pressure at the gas revovery plant in Ämmässuo between 1996 and 2004 and optimized (as in Ämmässuo) between 2002 and 2005 in Kujala, which increased the methane recovery efficiencies in both landfills. For example with k of 0.05 and L_o of 110 m^3/t methane collection efficiency increased from 20 % (in 1996) to 122 % (in 2006), whereas with k of 0.07 and L_o of 110 m^3/t methane recovery increased from 16 % (in 1996) to 101 % (in 2006) and with the previously (in this study) determined k (0.17) and L_o (110 m^3/t) gas recovery increased from 9 % (in 1996) to 75 % (in 2006) in Ämmässuo (Table 8). In Kujala gas recovery efficiency with k of 0.03 and L_o of 30 m^3/t increased from 79 % (2002) to 102 % (2005), whereas with the k of 0.06 and L_o of 30 m^3/t methane recovery efficiency increased from 61 % (in 2002) to 81 % (in 2005) and with the present determined k of 0.05 and L_o of 30 m^3/t recovery efficiency increased from 64 % (in 2002) to 85 % (in 2005) in Kujala.

The methane generation rates were also studied using higher L_o (from 130 to 170 m^3/t and from 40 to 60 m^3/t in Ämmässuo and Kujala, respectively) than determined in the present waste characterisation study, with the experimentally determined k (of 0.17 for Ämmässuo and 0.05 for Kujala) (Table 8). The methane recovery efficiencies in Ämmässuo then varied from 8 % (in 1996) to 63 % (in 2006) with L_o of 130 m^3/t, while with L_o of 170 m^3/t the methane recovery efficiencies varied from 6 % to 48 %. In Kujala methane recovery efficiencies varied from 50 % (in 2002) to 63 % (in 2005) with L_o of 40 m^3/t, while with higher L_o (60 m^3/t) the methane recovery rates varied from 36 % (in 2002) to 49 % (in 2005).

4.3.3 Total methane generation potential

The total methane generation potential of the two landfills during their lifetime was modeled using experimentally determined L_o (110 m^3/t for Ämmässuo and 30 m^3/t for Kujala) and also higher L_o (130 m^3/t for Ämmässuo and 40 m^3/t for Kujala), as some degradation was evident prior to sampling for the experimental analysis of L_o in both landfills. The modeled total methane generation varied from 984 (L_o of 110 m^3/t) to 1 163*10^6 m^3 (L_o of 130 m^3/t) and from 108 (L_o of 30 m^3/t) to 144*10^6 (L_o of 40 m^3/t) in Ämmässuo and Kujala, respectively (Table 9). k has no effect on the total methane generation, but the time needed to achieve a certain proportion of the total methane generation potential is determined by k. Thus the k determined in the present study by the experimental data (0.17 for Ämmässuo and 0.05 for Kujala) were used in the models for both landfills. The results showed that, e.g., approximately 90 % of the total methane generation potential will be achieved in Ämmässuo in 2013 (k of 0.17) and in Kujala in 2020 (k of 0.05). Assuming that 85 % of the generated methane will be recovered the remaining (from 2006 onwards) recoverable methane potentials are 319*10^6 m^3 (L_o of 110 m^3/t) and 378*10^6 m^3 (L_o of 130 m^3/t) with k of 0.17 in Ämmässuo and 45*10^6 m^3 (L_o of 30 m^3/t) and 59*10^6 m^3 (L_o of 40 m^3/t) with k of 0.05 in Kujala. Thus during the lifetime of the landfills the proportion of recovered methane will vary from 47 % (L_o of 110 m^3/t) to 44 % (130 m^3/t) with k of 0.17 and from 33 (L_o of 30 m^3/t) to 40 % (40 m^3/t) with k of 0.05 of the total methane potential in Ämmässuo and Kujala, respectively (Table 9, Fig. 15).

The proportion of methane emissions was calculated by subtracting the proportion of the total recovery potential (461 and 510*10^6 m^3 in Ämmässuo; 43 and 48*10^6 m^3 in Kujala) from the total methane generation potential (984 and 1 163*10^6 m^3 in Ämmässuo; 108 and 144*10^6 m^3 in Kujala) (Table 9). The total methane emission potentials are 523*10^6 (L_o of 110 m^3/t) and 653*10^6 m^3 (L_o of 130 m^3/t) in Ämmässuo as well as 65*10^6 (L_o of 30 m^3/t) and 96*10^6 m^3 (L_o of 40 m^3/t) in Kujala.

TABLE 9 The total and remaining methane potentials as well as recovery potentials by varying L_o (and k) of Ämmässuo and Kujala landfills (IV).

	Ämmässuo (*10^6)		Kujala (*10^6)	
Actual cumulative methane recovery	190 (from 1996 to 2006)		6 (from 2002 to 2005)	
	L_o = 110 m^3/t and k = 0.17	L_o = 130 m^3/t and k = 0.17	L_o = 30 m^3/t and k = 0.05	L_o = 40 m^3/t and k = 0.05
Total methane generation potential	984	1 163	108	144
Total recovery potential[1]	461	510	43	48
Total methane emission	523	653	65	96
Methane emissions before implementation of gas recovery[2]	185	219	55	73
Remaining methane generation potential since 2006 (Ämmässuo) and 2005 (Kujala)	319	378	45	59
Remaining methane emissions[3]	48	57	7	9

[1] Actual recovery + modeled methane generation by recovery rate of 85 % of the generated methane since 2006 in Ämmässuo and since 2005 in Kujala; [2] Gas recovery implemented in 1996 in Ämmässuo and in 2002 in Kujala; [3] From 2006 onwards in Ämmässuo and from 2005 in Kujala, assuming that 85 % of the generated methane is recovered (methane oxidation not considered)

The remaining (from 2006 onwards) methane emission potentials are 48*10^6 (L_o of 110 m^3/t) and 57*10^6 (L_o of 130 m^3/t) with k of 0.17 in Ämmässuo, while in Kujala the remaining methane emissions are 7*10^6 m^3 (L_o of 30 m^3/t) and 9*10^6 (L_o of 40 m^3/t) with k of 0.05, if 85 % of the generated methane is recovered in both landfills. The proportions of methane emissions before the implementation of methane recovery were 19 % (L_o of 110 m^3/t and 130 m^3/t) and 41 % (L_o of 30 m^3/t and 40 m^3/t) of the total methane generation potentials in Ämmässuo and Kujala, respectively. The major (81 %) proportion of the methane emissions was generated before the implementation of the gas recovery system in Kujala, while the corresponding proportion was minor (32-34 %) in Ämmässuo.

4.4 Characterisation of leachate and gaseous emissions from initial phases of landfilling mechanically and mechanically-biologically treated municipal solid waste residuals (V)

4.4.1 Temperature

The vertical temperature profile in the mechanically and mechanically-biologically treated MSW residuals was monitored at two sites in both lysimeters. During the study period, the temperatures ranged from 8 to 29 °C and from 5 to 23 °C in the M and MB lysimeters, respectively (Fig. 16), while the ambient temperature ranged from -19 to 27 °C. The highest (21-26 °C) temperatures were detected at the beginning of the landfilling (December 2003) in the M residual, after which temperatures remained at approximately the same level in both residuals until towards the end of the study period (June 2005 onwards), when the temperature in the M increased more than in the MB lysimeter. In both lysimeters the temperature in the highest stratum (0.5-0.8 m) corresponded most clearly to the ambient temperature. During the summers of 2004 and 2005 (from June to October) the top layer of the MB landfill was totally covered by vegetation, while the M landfill remained almost free of vegetation. The vertical temperature profile in the mechanically and mechanically-biologically treated MSW residuals was monitored at two sites in both lysimeters. During the study period, the temperatures ranged from 8 to 29 °C and from 5 to 23 °C in the M and MB lysimeters, respectively (Fig. 16), while the ambient temperature ranged from -19 to 27 °C. The highest (21-26 °C) temperatures were detected at the beginning of the landfilling (December 2003) in the M residual, after which temperatures remained at approximately the same level in both residuals until towards the end of the study period (June 2005 onwards), when the temperature in the M increased more than in the MB lysimeter. In both lysimeters the temperature in the highest stratum (0.5-0.8 m) corresponded most clearly to the ambient temperature. During the summers of 2004 and 2005 (from June to October) the top layer of the MB landfill was totally covered by vegetation, while the M landfill remained almost free of vegetation.

4.4.2 Composition of pore gases

The composition of pore gases at different depths were characterised in order to determine stages of degradation and in the M and MB residuals. Among the pore gases in the landfill body O_2 was detected mainly at the depth of 0.5 m in the MB lysimeter and also at 1.0 m in the MB lysimeter and 0.5 m in the M lysimeter (Fig. 17).

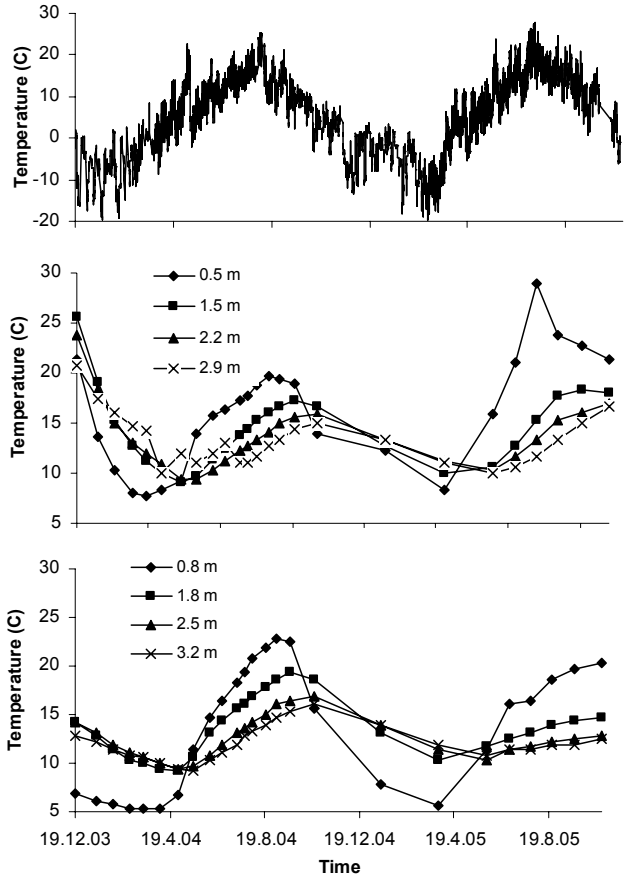

FIGURE 16 Ambient temperature (top) and temperature in M (middle) as well as in MB residual (bottom) lysimeters at four different depths. Depths were measured from top of the waste body (V).

The CH_4 concentrations were higher in the MB than M lysimeter, except in the latter part of the study at depth of 0.5 m, while the CO_2 concentration was mostly higher in the M than MB lysimeter. After one year of landfilling (October 2004) the CH_4 and CO_2 concentrations at depth of 1.5 m were 47-54 % and 33-34 % in the MB lysimeter, while in the M lysimeter the respective values were 12-16 % and 46-53 %.

4.4.3 Leachate generation

Leachate flow from the M and MB residuals was measured in order to characterise leachate generation and determine leachate solids (L/S) ratio with respect to precipitation. The leachate flow increased when the snow started to melt due to increasing ambient temperatures in April 2004 (Fig. 18).

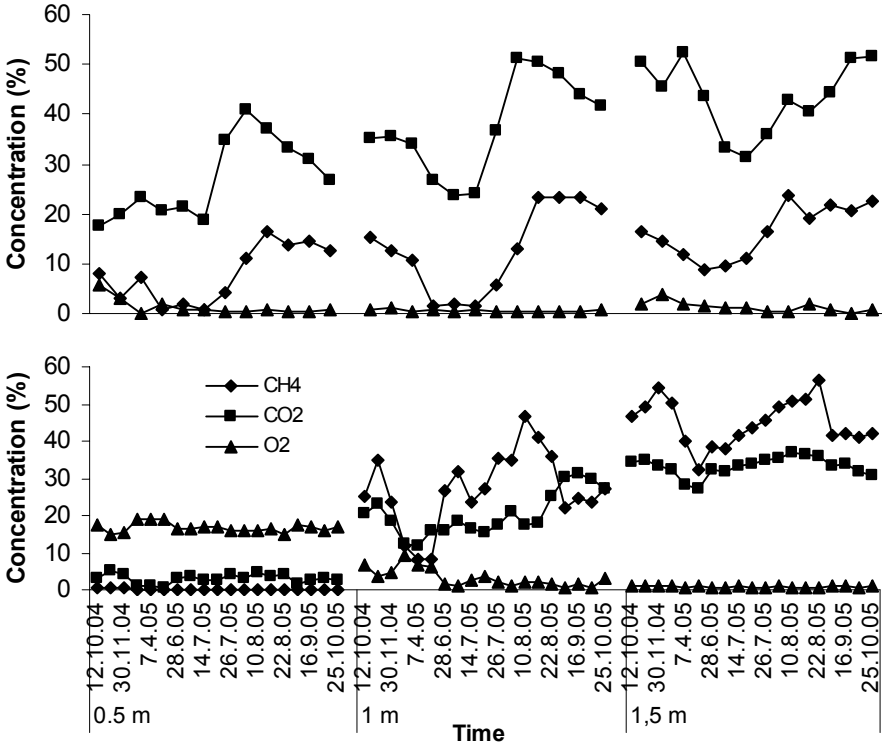

FIGURE 17 Pore gas concentrations at different depths of M (top) and MB residuals (bottom) (V).

During the study the highest daily leachate flows were about 0.7 m^3 and 0.9 m^3 (5.4.2005), while the monthly flows varied from 0.6 to 2.4 m^3 and from 0.4 to 3.2 m^3 in the M and MB lysimeters, respectively (data not shown). The cumulative leachate flows during the 426 d period before leachate recirculation in the MB lysimeter were 19.2 and 16.2 m^3 from M and MB lysimeters, respectively, corresponding to 70 and 59 % of the cumulative rainfall (27.3 m^3). During the leachate recirculation period (140 d from June to October 2005) 8.9 m^3 (309 mm as precipitation, weekly 12-37 mm) leachate was recirculated back to the MB landfill body and 1.3 m^3 leachate was discharged from the MB lysimeter, while from the M residual the discharged flow during the same period was 3.4 m^3. At the end of the study (640 d) total cumulative leachate flows to the outside of the lysimeters were 25.3 m^3 and 19.4 m^3 (63 % and 48 % of rainfall) from M and MB lysimeters, respectively. The obtained annual leachate flow/solid (L/S) ratios, in 2004) in the landfill bodies were 0.24 and 0.20 in M and MB, respectively, without leachate recirculation, while leachate recirculation increased the annual L/S-ratio in the MB body to 0.30 in 2005.

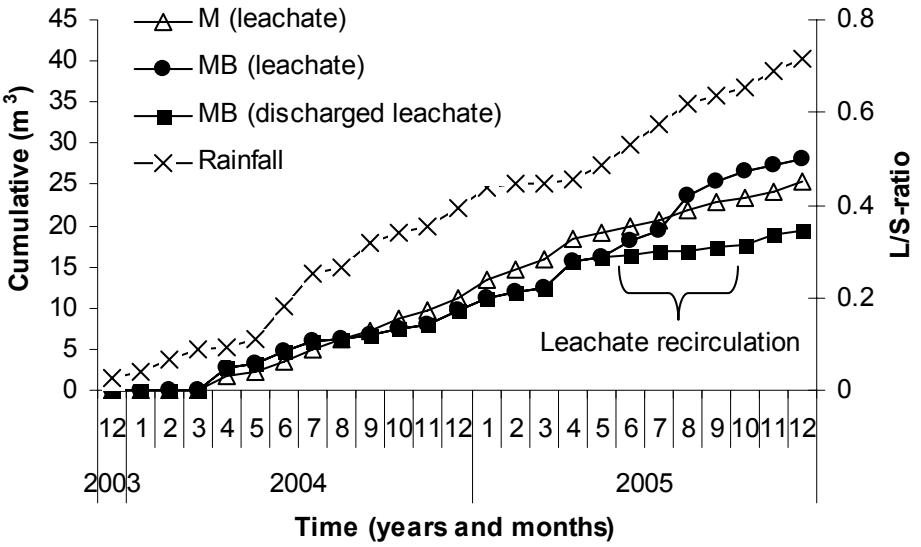

FIGURE 18 Cumulative leachate volumes and leachate/solids (L/S) ratios of M and MB residuals and rainfall in 2004-2005. The discharged leachate differs from leachate volume in MB residuals, due to leachate recirculation (V).

4.4.4 pH, redox and conductivity in leachate

pH, redox potential and conductivity in the leachate were determined in order to characterise stages of degradation and leachate quality in the M and MB residuals. The pH of the M leachate varied from 5.8 to 6.1 during the study, while the pH of the MB leachate decreased from 7 to 5.5 after one month of landfilling for the following two weeks (Fig. 19). Afterwards pH of the MB leachate increased to 6.8, and remained thereafter at 7-8 with increasing values after the summer season (September-October). The redox was throughout higher in the M (-200-0 mV) than MB leachate (-300--200 mV) and increasing values were observed after August in 2004 and 2005, and occasionally values above 0 mV in the M leachate. The M leachate had constantly higher conductivity (approximately 3000 mS/m) than the MB leachate (approximately 1000 mS/m). However conductivity showed a decreasing trend in both leachates during the study period.

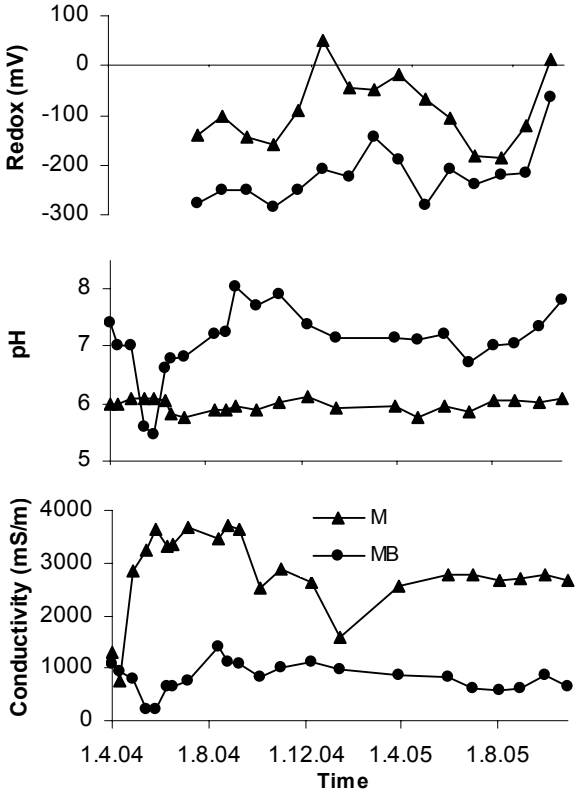

FIGURE 19 Redox-potential (top), pH (middle) and conductivity (bottom) of M and MB leachates (V).

4.4.5 Leaching of NH_4-N and SCOD

Leaching of NH_4-N and SCOD was determined in order to determine leachate load and characterise the degree of stabilisation of nitrogen and organic matter obtained by the mechanical and mechanical-biological treatments. NH_4-N (filtered samples) concentrations were 5-10-fold higher in the M (approximately 600-1800 mg/l, mean 1162 mg/l) than MB leachate (approximately 100-400 mg/l, mean 258 mg/l), where lower concentrations were found towards the end of the study period (Fig. 20). In both leachates the concentrations of NH_4-N and its proportion of nitrogen content (NH_4-N/TKN ratio, TKN non-filtered samples) increased during the beginning of the study (until November 2004), more in the M leachate, thereafter the concentrations and NH_4-N/TKN ratios varied without clear trends (Fig. 21). The NH_4-N/TKN ratios were, excluding some individual samples, 40-70 % and 70-90 % in the M and MB leachate, respectively. During the period (426 d) before leachate recirculation the leaching of NH_4-N was about 5-fold more from the M (391 g/t TS) than MB residual (79 g/t TS), and the leaching of TKN about 7-fold more from the M (694 g/t TS) than MB residual (103 g/t TS) (Fig. 20).

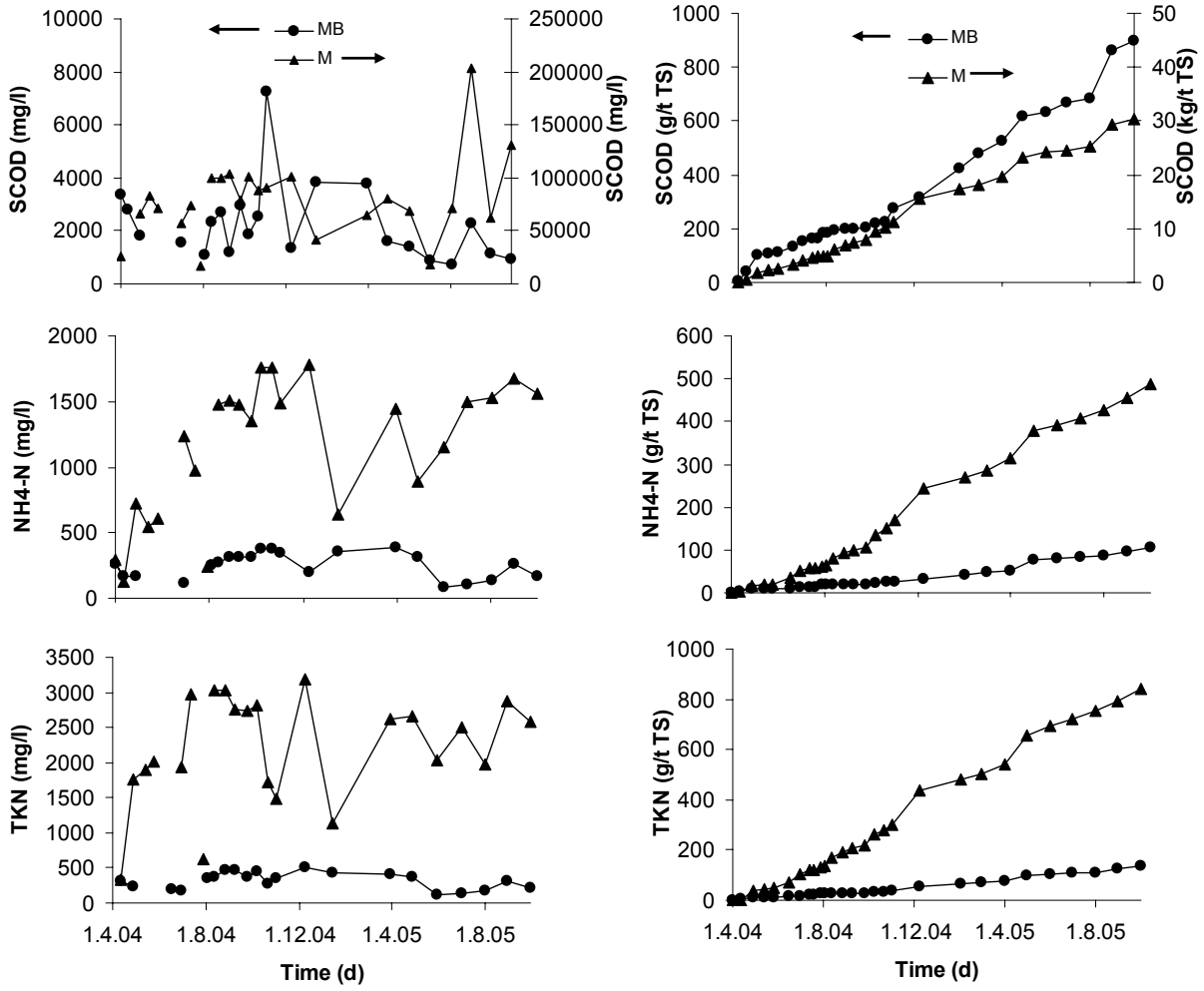

FIGURE 20 SCOD (top), NH_4-N (middle) and TKN (bottom) concentrations (mg/l) and leachate loads (g/t TS or kg/t TS) of M and MB residuals. Leachate recirculation was practised during 140 days, from 8th of June till 24th of October 2005 (V).

In the M leachate the SCOD values ranged from ca 20 to 100 g/l peaking at 200 g/l while in the MB leachate the SCOD values were initially typically 2-4 g/l levelling down to 1-2 g/l towards the end of the study period (Fig. 20), although the SCOD load in the MB leachate increased slightly at the end of study. During the period (426 d) before the leachate recirculation (in MB residual) approximately 40 times more SCOD was leached from the M (24.2 kg/TS) than MB residual (0.6 kg/t TS).

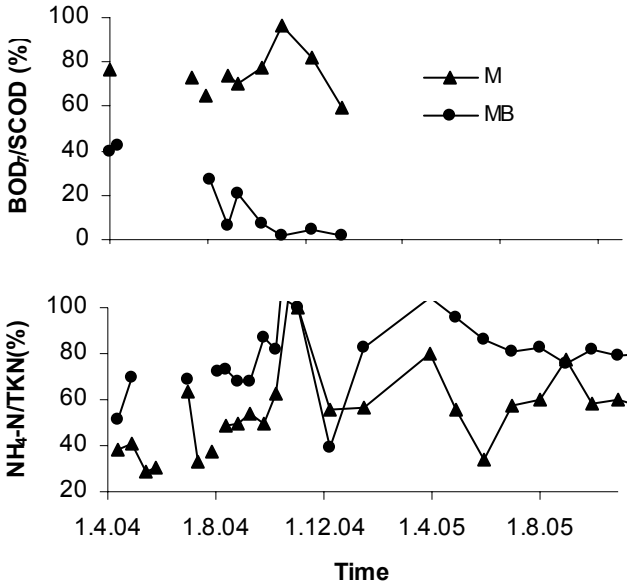

FIGURE 21 BOD_7/SCOD and NH_4-N/TKN ratios of M and MB leachates (V).

4.4.6 Characterisation of organic matter in leachate

BOD_7 and VFA in the leachates were determined for selected samples in 2004 and BOD_7 once in both leachates in 2005 in order to characterise the stages of degradation and level of stabilisation during landfilling of M and MB residuals. In the M leachate the BOD_7/SCOD- (Fig. 21) and the VFA_{cod}/SCOD-ratios (Fig. 22) were 59 – 96 % and 19-91 %, respectively, while in the MB leachate the ratios decreased from the initial ~40 % to less than 7 % for BOD/SCOD in 6 months and from the initial ~48 % to under 6 % for VFA_{cod}/SCOD in a month and remaining thereafter at < 1 % and peaking up to 15 % after three months. The BOD values were 24 g/l in the M and about 0.05 g/l in the MB leachate after 10 months' (January 2005) leachate flow, while the respective SCOD values were 41 g/l and 3.8 g/l. The total VFA concentrations in the M leachate varied from 3.5 to 25 g/l and in the MB leachate from 0.009 to 2.7 g/l.

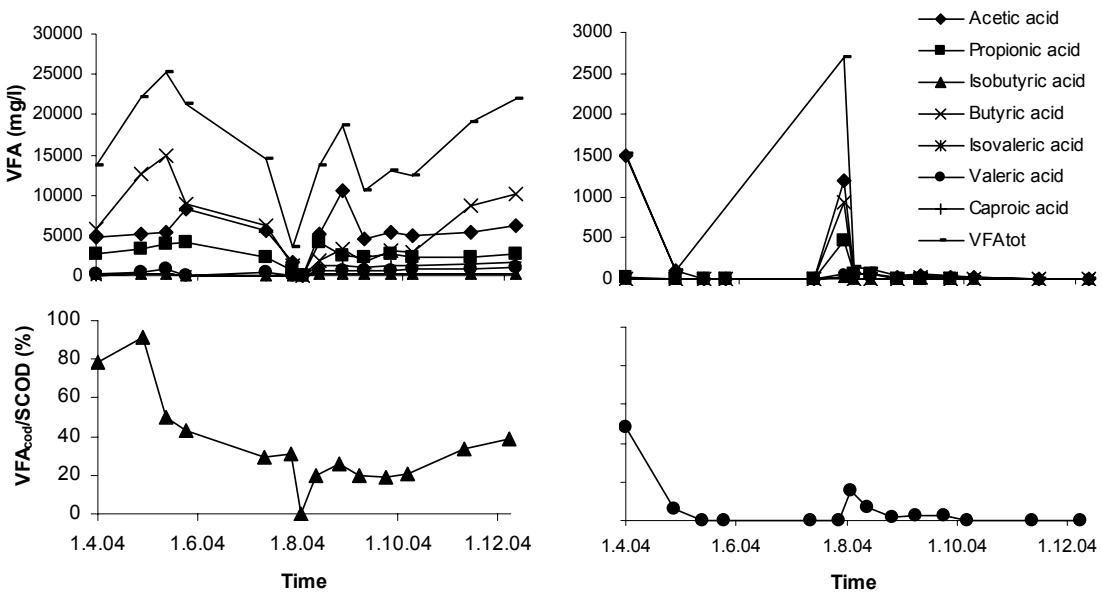

FIGURE 22 VFAs (top) and VFA_{cod}/SCOD ratios (bottom) in M (left) and MB (right) leachates (V).

4.4.7 Carbon dioxide, methane and nitrous oxide emissions

Gas emissions were measured in order to determine the gas generation rate and its composition. Carbon dioxide formed the major proportion of the gas emissions, ranging from 15 to 71 l/m^2 d (mean of separate measurements 43.7 l/m^2 d) and from 5 to 41 l/m^2 d (mean 22.4 l/m^2 d) in the case of the M and MB residuals, respectively (Fig. 23). Both methane and nitrous oxide emissions were higher from the M than MB residuals; range from 0.05 to 12.4 l CH_4/m^2 d (mean 1.9 l CH_4/m^2 d) and 0.001-0.14 l N_2O/m^2 d (mean 0.025 l N_2O/m^2 d) in M; range from <0.02 to 0.3 l CH_4/m^2 d (mean 0.08 l CH_4/m^2 d) and <0.001 to 0.05 l N_2O/m^2 d (mean 0.01 N_2O l/m^2 d) in MB.

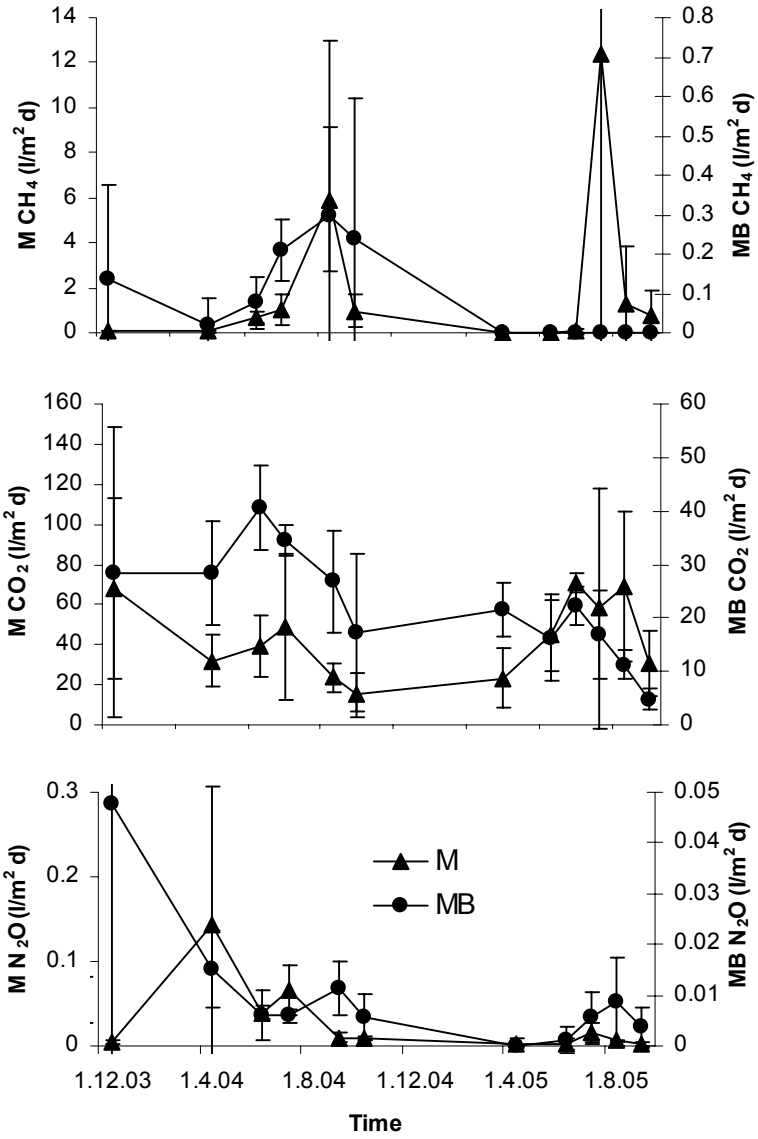

FIGURE 23 The CH_4 (top), CO_2 (middle) and N_2O (bottom) emissions of M and MB residuals (V).

5 DISCUSSION

5.1 Monitoring of landfills by waste and leachate characterisation

The present study, which attempted to characterise landfill bodies approximately 20 and 50 years old shows the difficulty attached to such an undertaking, but also provides some important information about the possibilities of characterising landfill contents and generalising the results to other landfills. Large variation in all the parameters studied was found in both landfills despite the fact that the samples were obtained by systematic vertical (3-4 heights) and horizontal (ca 50 x 50–100 m) sampling and that the initial sample size was relatively large (300-500 l in Ämmässuo, 10-20 l in Kujala) compared to the procedures used in some previous landfill studies (e.g., 0.5-1.0 l, Bookter & Ham 1982; ca 1 l, Östman et al. 2006; ca 100 l, Suflita et al. 1992). It is evident therefore that characterisation of a heterogeneous landfill body requires intensive sampling (this study, Bookter & Ham 1982, Ham et al. 1993, Suflita et al. 1992) and that the minimum number of sampling points varies according to parameters studied, landfill properties and the accuracy required - thus general recommendations on the minimum number of samples cannot be given. However in order to study vertical profile at least from 3 to 4 sampling depths would be needed at each sampling point in landfills 30 m deep to characterise vertical layers, as was done in this study. At least one of the sampling depths should be below the water table (if high water table exist) where the waste is usually oldest and high moisture content and leachate movement favour stabilisation, as shown by Hartz and Ham (1983). In present study the batch assays showed that water addition may speed up methane production, which is likely due to the dilution of inhibitory compounds or/and better transport of substrates to microorganisms. However, as expected, the major stimulation to methane production in the batch assays was achieved with the addition of inoculum, which in practical conditions has been considered by adding inoculum to recirculated leachate (Bae et al. 1998).

In general, the two landfills studied were very different, despite the facts that both were major MSW landfills in their regions, both received waste from similar communities, and both had similar climatic conditions in close proximity to each other (110 km). In Kujala the longer stabilisation time (around 50 years) and greater amount of inert material as well as landfill practices had resulted in a low VS/TS ratio in the bottom layer. On the other hand, in Kujala, the present VS/TS (26 %) was higher than the 20 % VS/TS found at the lowest 12 m in the late 1980s (Ettala et al. 1988), which could suggest that leaching of organics from the upper layers may increase VS/TS ratio in bottom layers. This was supported also by the fact that the leaching of SCOD showed only a slight depth-related decrease, while the VS/TS ratio decreased more significantly (from top to bottom) in Kujala. In the present study a VS/TS ratio of 16 %, indicating well stabilised waste was found only in the bottom layer in Kujala. The fact that not only biodegradable (e.g., cellulose), slowly biodegradable (e.g., lignocellulose) and recalcitrant material (e.g., plastics), but also inert material (e.g., ash, soil) contributes to the sample's TS (and VS) and caused the high variation in VS/TS ratios, also at the lowest layers in both landfills. In Ämmässuo, the 3-fold higher BMPs in middle layer than in two middle layers in Kujala were apparently due to differences in landfilled waste characteristics, as the age of the waste in these layers was similar in both landfills. The highest BMPs (160-180 m^3/t TS) in Ämmässuo were approximately half that of food waste (301 m^3/t TS, Eleazer et al. 1997), and higher than reported for grey waste (46-101 m^3/t TS, Jokela et al. 2001, Jokela et al. 2002), suggesting high methane potential despite the fact that the waste was mainly from a period (1995 onwards) when source segregation was increasingly being implemented in this region. Furthermore, variation in all the parameters studied was clearly larger in Ämmässuo than in Kujala. These differences could indicate that waste, especially in the middle and top layers in Ämmässuo contained significant amounts of biodegradable material which was probably just beginning to degrade. Moreover BMP, VS, VS/TS, NH_4-N and COD leaching were higher – some even several-fold - in Ämmässuo than in Kujala, while TS was lower in Ämmässuo than in Kujala. In both landfills pH was within a range (6.7 to 8.9 and 7.0 to 8.2 at Ämmässuo and Kujala, respectively) enabling methanogenesis, although a smaller range (6.8-7.4) has been determined for optimum methanogenesis (Barlaz et al. 1990). The lower pH in the top layers of landfills (Ämmässuo; Jones et al. 1983) is apparently due to the accumulation of organic acids on account of the low populations of methanogenic bacteria, which can not utilize the acids at same rate as they are produced in fresh waste (Barlaz et al. 1990).

However both landfills had some common features, e.g., high proportions of soils or soil-like materials (including ash). These were, apparently from daily soil cover, ash, and demolition waste. There were similar proportions of wood at both sites (l5-16 % in Ämmässuo and 9-13 % in Kujala); other studies have indicated the recalcitrance of wood under landfill conditions (Gardner et al. 2003, Micales & Skog 1997). In both landfills the proportion of paper and cardboard decreased towards the bottom of the landfill, indicating their degradation. This is in accordance with the fact that their main components are cellulose and

hemicellulose, which have been found to contribute up to 91 % of the methane produced in landfills (Barlaz et al. 1989). Similar contents of plastics (5-9 %) were found in all layers in both landfills, except in the bottom layer of Kujala, where its low content (1 %) might be due to either low original percentages or on-site incineration at time of landfilling.

Besides waste characterisation the present study shows the feasibility of leachate sampling to characterise leachate quality and stages of degradation within a MSW landfill that has been in operation for several decades. In particular, it seems that leachate quality can vary considerably both vertically and horizontally, as found in the present samples from monitoring wells located at a distance of only 5 to 150 m from each other in the Kujala landfill. It is therefore evident (Kjeldsen & Christophersen 2001, this study) that reliable characterisation of leachate quality within a MSW landfill requires the systematic installation of monitoring wells for sampling in saturated waste layers and water table measurements. Moreover, waste quality in different areas and layers should be considered in planning the placement of wells as also should the lengths and screen range of wells to take into account the effect of attenuation (e.g., dilution or degradation) processes on leachate quality. The minimum number of monitoring wells would seem to depend on the properties of the landfill (waste age and landfill filling procedure) as well as on the aim of the sampling, and thus no general recommendations can be given. It was not possible to differentiate and observe the short-term effects of leachate recirculation on internal leachate quality in the present 50-year-old MSW landfill, despite the fact that leachate quality varied most in the monitoring wells in close proximity to the leachate recirculation canals. This may be because the variation in leachate quality due to leachate recirculation is low and /or because the variation in leachate quality is affected by other factors such as degradation processes and water flows due to precipitation and/or melting snow.

In the studied landfill the leachate at the bottom (up to 2 - 3 meters from the bottom) was strongly (3-8 fold) diluted compared to that at higher levels (from 4 to 7 meters from the bottom) as indicated, e.g., by the fact that the COD value and chloride concentration decreased towards the bottom. The decrease in COD values alone could indicate methanisation of COD with landfill body depth, while chloride is assumed to be useable as a tracer compound due to its inert nature (Rosqvist et al. 2005, van der Sloot et al. 1999). The mean COD value (5610 mg/l) and the mean NH_4-N concentration (1732 mg/l) obtained from the higher level monitoring wells were higher than the reported means for landfills from 1 to 5 years old (3810 mg/l for COD and 405 mg/l for NH_4-N, respectively; Kruempelbeck & Ehrig 1999). Thus it is clear that the present landfill still contains high amounts of easily transportable organic material and nitrogen, which flow through the landfill. The BOD/COD ratio (mainly 0.05-0.2) in most of the observation wells was typical of a methanogenic (0.02-0.24, Kjeldsen et al. 2002) landfill, indicating the degradation of organic matter within the landfill.

5.2 On-line monitoring of water table, temperature and leachate quality in landfills

This study shows the feasibility of the on-line monitoring of electrical conductivity and pH in internal leachate in a MSW landfill. The values obtained from the on-line measurements were rather similar to those obtained by the sampling and field measurements. The differences between the on-line and field meter values were systematic. The field meters showed higher values for electrical conductivity and pH than the on-line measurements. The lower values obtained from the on-line monitoring could be caused by landfill gas flow through leachate as landfill gas bubbles in leachate may decrease electrical conductivity, while release of carbon dioxide dissolved in leachate tends to increase the pH of samples immediately after sampling. The variation in the on-line measurements of electrical conductivity may indicate changes in internal leachate flow or characteristics as this variation coincided with the leachate recirculation in a monitoring well in Kujala where the electrical conductivity of the recirculated leachate was lower (range mainly 300 - 500 mS/m) than the internal leachate as a whole (mean approximately 1500 mS/m). However, a similar variation in electrical conductivity was not detected by field measurements performed approximately once in month; thus on-line monitoring seems to be a more accurate method of monitoring short-term variation in electrical conductivity.

In general, the on-line measurements indicated that the changes in the water table were low even during a monitoring period as long as one and a half years, while some gradual and temporary changes took place in temperature and electrical conductivity in some monitoring wells. Thus the usability of on-line measurements is highly case- and target- dependent. However, monitoring of water table and temperature has been considered important, especially when operating a landfill as a bioreactor, due to fact that rise in the water table may cause increasing leaching into groundwater and a high temperature can damage the lining, leachate and gas collection systems (Benson et al. 2007). Finnish legislation (Finnish Government 1997) requires monitoring of the water table in the internal leachate and the temperature within the landfill as well as the electrical conductivity in the external leachate. However, on-line monitoring is often not thought to be necessary, despite the fact that, as stated in the legislation (Finnish Government 1997), it is necessary to ensure that the various processes of degradation proceed as intended and that environmental protection structures (e.g., drainage layer, liner and leachate recirculation structures) are fully functional. Moreover it is important that monitoring is systematic, samples are representative and that changes in leachate quality are noted quickly. Thus the variation observed in electrical conductivity (present study) also demonstrated the difficulty of representative manual sampling as, for example, the concentration of ammonium nitrogen, which has been generally considered one of the main pollutants in MSW leachate (Kjeldsen et al. 2002), has shown tendency to correlate with electrical conductivity (Marttinen et al. 1999).

In both landfills the highest temperatures (35 °C and 39 °C) approximated to those previously determined (reviewed by Barlaz et al. 1990) in the mesophilic range (34-42 °C) for optimal methane generation. It is clear that temperatures in a landfill are likely to vary in different parts and layers, as shown by the range from 5 to 39 °C previously measured in Ämmässuo (Sormunen et al. 2004). The present study suggests that variation in the temperature of the internal leachate is in part due to the height of the landfill, at least in landfills located in a boreal climate. In the present on-line measurements marked changes were not observed in internal leachate temperature in either landfill during the one and a half year study period, despite the -28 to 30 °C annual variation in ambient temperature. Thus landfill gas production in the saturated zone of these landfills is likely to continue without showing significant seasonal variation.

Technically, the monitoring systems investigated in this study appeared, in both landfills, to work without any major problems. However the electrodes need regular maintenance and further need for calibrations may exist, particularly in the longterm. It is difficult on the basis of the present study to generalize regarding minimum calibration intervals as it has previously been reported (Thomas and Pouet 2005) that the function of on-line measurements is highly dependent on wastewaters characteristics, e.g., the wastewater properties causing fouling of electrodes. It seems that maintenance (including cleaning and calibration) intervals between 1 and 2 months for electrical conductivity and pH might be appropriate in the landfill studied here. Contact between landfill gas and electronics should be avoided when implementing measurements, as a previous study (Kim et al. 2005) has shown that a major proportion of the sulphur in landfill gas exists in the form of hydrogen sulphide, which is highly corrosive. In addition to corrosion, settling of the landfill may cause some movement or bending in the monitoring well; thus monitoring wells with an inner diameter of >50 mm are highly recommended to prevent electrodes sticking and facilitate maintenance. Moreover foaming of the leachate in monitoring wells was seen in both studied landfills. This can cause inaccuracy in water table monitoring, and thus monitoring wells less prone to foaming are preferable for the purpose of monitoring.

5.3 Determination of methane generation rate and potential in municipal solid waste landfills

The present results demonstrated that the methane generation rate (k) and (L_o) in the two Finnish landfills studied are apparently different, even if the landfills are located at distance of approximately only 100 km from each other and share similar climatic conditions (e.g., mean annual precipitation 601-700 mm).

The L_o of landfilled waste (as bulk waste until the 1990s and within the last decade increasingly as a source-segregated grey waste) seems to be approximately 130 m^3/t (wet) in Ämmässuo and approximately 40 m^3/t (wet) in Kujala.

These values are suggested by the experimental determination of L_o and comparison of the modelled methane generation with the amounts of methane actually recovered in the last 2-3 years, when the gas recovery system has been fully implemented and both landfills are close to their final heights. The actual original methane generation potentials of the landfilled waste was probably higher than the L_o experimentally determined from the waste samples taken from the upper layers of the landfill bodies, in which waste had been landfilled within the 2-3 years before sampling in both landfills. This, is likely to be the case as it has been assumed (e.g., IPCC 2006) that methane production is highest in the first few years of landfilling due to degradation of easily degradable materials. On the other hand easily degradable materials may induce the acidogenic phase of degradation, when methane production is limited, as a lag of approximately 2 years in methane production was considered typical in US landfills, and even in so called wet landfills where moisture content is specified by a bioreactor landfill operation (Faour et al. 2007).

The experimentally determined *k* values (*k* of 0.17 for Ämmässuo and 0.05 for Kujala) indicated a much higher rate of degradation in Ämmässuo than in Kujala. This was probably caused by the higher proportion of easily degradable waste, such as kitchen biowaste, as it has been found that the biodegradable fraction can account for as much as 70 % (of which 25.2 % may consist of kitchen biowaste) of the landfilled household waste in Ämmässuo (YTV 2004), where as in Kujala the corresponding figure was 60 % (PHJ 2006). For comparison a default *k* of 0.06 (range 0.05-0.08) is used in national greenhouse gas inventories (IPCC 2006) for the degradation of rapidly degrading waste such as food waste in dry temperate conditions (conditions determined by the ratio of mean annual precipitation/potential evapotranspiration, MAP/PET<1), while in wet temperate conditions (MAP/PET>1) approximately the same *k* (range 0.1-0.2, default 0.185) is used for food waste as that proposed in the present study (0.17) for landfilled bulk waste in Ämmässuo. On the other hand the present *k* of 0.17 for methane generation in Ämmässuo was lower than the estimated *k* of 0.3 by Faour et al. (2007) for recovered methane in wet (specified moisture by leachate recirculation and moisture addition) landfill conditions. The USEPA defines a bioreactor landfill according to a moisture content of 45 % (wet weight) in landfills where leachate has been used for moisture addition (Reinhart et al. 2005). Previously (Faour et al. 2007) *k* was determined for methane recovery, whereas present to study determined *k* for methane generation, and thus it seems that a *k* of 0.17 may be appropriate for Ämmässuo. Moreover the high moisture (mean moisture content of 46 %, Sormunen et al. 2008) in Ämmässuo may favor biodegradation as higher *k* (from 0.3 to 0.5) has been reported for wet landfills in previous studies (Faour et al. 2007, Yazdani et al. 2006). In Kujala the determined *k* of 0.05 is same as the default *k* (range 0.04-0.06) used by IPCC (2006) for moderately degrading waste such as garden waste in dry temperate conditions (MAP/PET<1). The fact that the present *k* of 0.05 determined in Kujala is much lower than that in Ämmässuo is probably caused by the fact that a larger part of the landfill body was more stabilized

during the several decades of landfill's operation, and thus the present *k* for bulk waste is low, although it is evident that with newer waste *k* will be higher.

The present results on modeling methane generation in landfills indicate the importance of the installation of a gas recovery system in determining the fate of the methane generated. In the two landfills studied here approximately 58 % (Ämmässuo) and 60 % (Kujala) of total methane generation during the landfills' history will be released without treatment, partly due to fact that approximately 19 % and 41 % of the total methane generation occurred in Ämmässuo (1996) and in Kujala (2002), respectively, before the installation of gas collection systems. Improved gas recovery efficiency (from 8 to 63 % in 1996-2006) has been obtained in Ämmässuo due to fact that the gas recovery has been implemented in larger areas and some areas have been covered by final cover structures.

5.4 Leachate and gaseous emissions from initial phases of landfilling mechanically and mechanically-biologically treated municipal solid waste residuals

The results of the present comparative lysimeter study clearly show that aerobic stabilization causes a marked reduction in gaseous and leachate emissions of mechanically processed municipal solid waste during the initial phase (25 months) of landfilling. The impact could be seen, e.g., in the 5-fold higher BMP, 5-10-fold higher SCOD values and 3-6-fold higher nitrogen concentrations in the M than MB leachate (similar leachate flows) and in the much smaller leachate loads from the MB than M residual, which were about 3 % in SCOD, 20 % in NH_4-N and 15 % in nitrogen. Furthermore, the greenhouse gas emissions, namely methane and nitrous oxide were up to 20-fold and 11-fold higher, respectively, from the M than MB residuals, despite the fact that the M residual was still mainly in the acidogenic phase. It is clear that the SCOD and nitrogen concentrations and loads from the initial phases of the landfilling of MB residuals are much lower than in the case of M residuals, and as decreasing trends were not observed it appears that the loads continue approximately at the same rates for a while (except SCOD from MB-residual) from both materials after the period studied here.

These differences in leaching of SCOD and nitrogen are suggesting the need for different types of leachate treatment concepts and technologies. On the basis of MB residual landfill simulation study the NH_4-N and organic matter pollution may require treatment as long as L/S ratio of 3 and 2.5 will be reached, respectively, which may take in landfill conditions 150-200 years depending on discharge limits (Höring et al. 1999). The present SCOD values (mainly from 710 to 3827 mg/l) in MB leachate were a little higher or at the same level as in a laboratory study (mainly from 500 to 2000 mg/l determined as COD, Leikam & Stegmann, 1997) and in a landfill lysimeter study (COD

from 762 to 3043 mg/l, Felske et al. 2003) or in a full-scale landfill study where the major part of the waste consisted of MB residuals (COD from 228 to 4670 mg/l, Bone et al. 2003). The present SCOD values in the M leachate varied from 16 000 to 203 000 mg/l and the values did not decrease during the study, which indicates a huge leachate pollution potential as the landfill conditions seem to remain strongly acidic (pH about 6) for at least two years. In MB landfills the acidogenic stage is commonly avoided on the evidence of the leachate quality (e.g., pH >7) found in a laboratory study (Leikam & Stegmann 1997), in a field-scale lysimeter study (Felske et al. 2003) and in a study of full-scale landfills (Bone et al. 2003) and the gas quality (CH_4/CO_2-ratio) found in a laboratory study (Bockreis & Steinberg 2005) as well as in a field-scale lysimeter study (Felske et al. 2003). The proportion of VFA_{cod} from SCOD was 19-91 % in the M leachate and mainly <1-15 % in MB leachate, indicating that proportion of easily degradable organic material is high in M leachate. The NH_4-N/TKN ratio increased in the MB leachate, indicating higher ammonification after 6 months of leachate flow, while the NH_4-N/TKN ratio was about 20 % lower in the M leachate. The nitrogen concentrations in the present study in the M leachate were about two thirds of the M leachate nitrogen concentration observed in other field study (Woelders & Oonk 1999) and about the same as in a laboratory study (Leikam & Stegmann 1997). The SCOD and NH_4-N concentrations in the MB leachate were slightly higher and nitrogen concentrations about same in the present study as in the corresponding laboratory study (Leikam & Stegmann 1997) in the first 1-3 months of the landfilling, while later, during the following three to five months, the NH_4-N and nitrogen concentrations increased in both studies.

The present lysimeter studies as well as previous experiences from laboratory studies (e.g., Cappai et al. 2005, Leikam & Stegmann 1997) and field studies show that M (Woelders and Oonk 1999) and MB residual landfills (Bone et al. 2003, Felske et al. 2003) have different mechanical and biological characteristics compared to each other as well as to untreated MSW. Because of the higher biological stability of MB, temperatures are lower in MB than M landfills and conventional MSW landfills. In present study temperatures were higher in M than MB residuals in the beginning of the landfilling and in the end of the study indicating higher biological reactivity in the M than MB residual. The densities of the waste bodies in this study were 0.9 t/m^3 and 1.0 t/m^3, while a density of 1.3 t/m^3 for a compacted M residual landfill (Woelders & Oonk 1999), densities of 1.0-1.6 t/m^3 for MB residual landfills (e.g., Felske et al. 2003, Scheelhaase & Bidlingmaier 1997, Stegmann et al. 2005) and densities of 0.5 - 0.8 t/m^3 typical for untreated waste landfills (Reinhart & Townsend 1998) have been reported. Due to higher density the water permeabilities are lower in MB landfills (10^{-5} - 10^{-10} m/s, Stegmann et al. 2005) than in untreated waste landfills (10^{-4} – 10^{-8} m/s, Reinhart & Townsend 1998), which may cause practical difficulties, e.g., reduced mechanical stability of the landfill body due to high pore water pressure (Stegmann et al. 2005). Moreover leachate recirculation may require a pressurised system when high recirculation rates (e.g., >30 mm per week) are used, as in the case of M residual (density 1.3 t/m^3) bioreactor landfill study (Woelders

& Oonk 1999). In the present study high leachate recirculation rates (up to 33-37 mm/week) were used into MB lysimeter (1.0 t/m^3), thus showing that lower compaction may enable leachate recirculation by a gravity-based system. However increased gas production was not observed due to leachate recirculation even though in the previous field-scale study (Lorber et al. 2001) the gas production was increased due to water addition in the MB residuals. In the present study the L/S ratio was increased by 0.11 in the MB-residual compared to the M-residual, which meant that L/S ratio rose by about 50 % of the annual L/S ratio (0.2-0.24) without leachate recirculation. Thus leachate recirculation can shorten the time needed to reach the discharge threshold value, e.g., for nitrogen as a previous laboratory study (Höring et al. 1999) have shown that leaching of nitrogen from the landfill in the long term is mainly determined by the L/S ratio. Moreover, in MB residuals the annual (2005) leachate discharge flow was reduced by about 30 % probably due to leachate recirculation and increased evaporation in the MB residual compared to the M residual.

The present results show that the methane emissions from the M and MB residuals were low (from 0.05 to 12.4 l/m^2d in M and from 0.02 to 0.3 l/m^2d in MB residuals) during the initial phases of landfilling compared to average methane emissions from whole MSW landfills (6-180 l/m^2d, as reviewed by Kettunen et al. 2006). In fact, the M residuals in present study had a high BMP (259 m^3/t TS) compared to the MB residuals (52 m^3/t TS), indicating that after the acidogenic stage, when the methanogenic stage commences, an increase of methane emissions occurs, and may require effective gas recovery and treatment. Based on the BMP assay, we estimated (using the CH_4 production by day 21, and on the assumption that methane contributed 60 % of the total gas production) that the mean GB21 value of the MB residual used in this study could have been 40-70 Nl/kgTS and not 20 Nl/kg TS as determined for one of the seven batches used in preparing the MB residual. This could indicate e.g. that some of the batches were not well stabilised in the reactor composting phase, as it has been reported (Binner & Zach 1999) that it is difficult to compensate failures during the first weeks of reactor composting by prolonged composting afterwards.

The fate and amounts of leachable compounds in landfills are also affected, aside from the landfill conditions themselves, by the biotic and abiotic factors that take place during the preceding processing/stabilisation stage. Aerobic stabilisation appears to decrease the organic (VS) more significantly than N content of M residual, as indicated by the lower VS/TS ratio of the MB residual (43 %) compared to the M residual (61 %), while the nitrogen content of the TS were about same in both residuals (0.5 % in M and 0.6 % in MB) in this study. In previous studies MB residuals contained approximately same amount (1.1 % Cabbai et al. 2005) or less (0.4 %, Boni et al. 2006) nitrogen (of TS) than M residuals (1.2 %, Cabbai et al. 2005, 1.0 %, Woelders & Oonk 1999) or nitrogen content remained the same (about 1 %) during biological stabilization (Heiss-Ziegler & Lechner 1999). These results (stable nitrogen content % of TS and loss of VS) mean loss of absolute nitrogen content during stabilisation, as part of nitrogen concentrates into a smaller waste mass. On the other hand, it has been reported

that absolute nitrogen content can be even more greatly reduced during aerobic stabilisation than in the present and some previous studies; e.g., Boni et al. (2006) reported that 15 days stabilized M residual contained 0.6 % nitrogen and after 90 days stabilisation the nitrogen content was reduced to 0.4 %. The mechanisms affecting the fate of nitrogen, however, are not fully understood, although a few possible mechanisms, which may affect the fate of nitrogen during MB treatment and landfilling of MB residuals can be suggested (Bone et al. 2003, Cappai et al. 2005). Part of the nitrogen is evaporated as NH_3 (18-1150 g/t treated waste), which can be recovered, e.g., by using scrubbers (Clemens & Cuhls 2003). Some NH_4-N (as well as organic material) may also be washed out in leachates from composts, thus requiring treatment. The amount of emitted non methane volatile organic compounds (NMVOC) as gas can rise to 600 g/ton (Soyez & Plickert 2002), methane from 6 to 8620 g/ton, carbon dioxide from 12 to 185 kg/ton and nitrous oxide from 1.44 to 378 g/ton treated waste (Clemens & Cuhls 2003) during the aerobic treatment of M residual. On the other hand part of the NH_4-N may also be nitrified during composting (Cappai et al. 2005, Heiss-Ziegler & Lechner 1999) and denitrified later in landfill conditions (Bone et al. 2003). Moreover composting seems to increase the proportion of humic nitrogen (Cappai et al. 2005, Heiss-Ziegler & Lechner 1999). For example, the proportion of humic nitrogen rose from about 17 % to over 40 % during six months composting (Ziegler 1997). It seems that the formation of humic nitrogen might be the major phenomenon, which explains the decreased ammonium nitrogen content and reduced nitrogen load emitted from MB-treated materials (Cappai et al. 2005). In fact in the present study the leaching of nitrogen and NH_4-N compared to the initial nitrogen content support the notion that nitrogen is strongly compounded - probably as humic nitrogen - into MB residual, as the proportion of leached nitrogen and NH_4-N of total values over 426 d were 1.7 and 1.3 %, respectively, from the MB and 14 and 8 % from the M-residuals.

5.5 Characterisation of landfills

In the present study several methods were used to determine landfill conditions and waste and/or leachate properties in order to characterise both old (approximately 50-and 20-year-old) landfills with unknown contents and new landfills with known contents of mechanically and mechanically treated MSW residuals (Table 10). The present results testify to the importance of landfill waste characterisation studies in seeking to determine the rate of degradation and current waste properties, e.g., water content, organic material, BMP and TKN. Internal leachate characterisation and on-line monitoring seem to be more feasible in the determination of current conditions, e.g., pH, BOD/COD ratio, water table and temperature in the leachate saturated zone of a MSW landfill. Moreover both waste and leachate characterisation studies are feasible methods

of monitoring degree of stabilisation and horizontal and vertical variation in waste quality and its leaching properties. In addition, characterisation of the landfilled materials is feasible on the basis of their organic matter and nitrogen budgets in landfills where the contents of mechanically and mechanically-biologically treated MSW are known.

In this study, the characterisation both of 50- and 20-year-old MSW landfills and of present/new landfills receiving mechanical or mechanical-biologically treated MSW was the primary objective. The generalisation of the information obtained to other landfills, even those with a similar social and waste management history can not be assumed as it appears that the data obtained is case-dependent. The present study shows that the different types of landfills studied can be characterised in according to their waste and leachate properties (Table 11). Some of the main characteristics found were:

- In the 50-year-old landfill (height approximately 25 m) well stabilised waste with low organic material content and BMPs were found in the bottom layers. In general the BMPs were lower (varying from 6-60 m^3/t TS) than in the 20-year-old landfill. The nitrogen content of the bottom layer was approximately 50 % of that in the top layers and the current highest rate of NH_4-N leaching was in the middle layers. Sealed bottom structures do not exist in landfills of this age, and thus a high water table can only exist if the natural soil under the landfill minimises the infiltration of leachate into the groundwater. A high water table favours biodegradation in the saturated zone.
- In the 20-year-old landfill (height approximately 25 m) BMPs varied greatly (from 1 to 183 m^3/t TS). BMPs were significantly lower at the bottom compared to middle and top layers in landfilled waste of different age. Leaching of organic materials was also lower in the bottom layer, which indicates a higher degree of stabilisation in bottom than top layers. NH_4-N leaching seems to occur from all layers, indicating thereby that the nitrogen load is longlasting. A high water table may exist, if the drainage layer or leachate collection fails to function properly. A high water table may enhance degradation in layers in the saturated zone, while low moisture content may limit biodegradation in the top layers.
- The landfill with mechanically treated MSW residuals contained high amounts of easily degradable organic materials, as indicated by COD content, which varied from 20 g/l to 100 g/l in leachate, and small particle size tended to promote the acidogenic phase (pH <6.1) of degradation, which may last over two years. During the acidogenic phase of degradation a high proportion of organic materials (24.2 kg COD/t TS) and ammonium nitrogen (391 g/t TS) leach out from landfills, while methane generation remains low (<16 %, methane content in pore gas) even if the BMP (259 m^3/t TS) indicates high methane generation potential. Thereafter methane production will increase; this may, require an effective system for the collection and treatment of landfill gas.

- The landfill with mechanically-biologically treated MSW was stabilised, especially with regarding to the leaching of organic matter (0.6 kg COD/t TS), compared to the landfills with untreated waste (means 6-19 kg COD/t TS) or mechanically treated waste (24.2 kg COD/t TS). The nitrogen content was higher (mean 0.6 % TS) than with untreated (mean 0.4 % TS in both landfills) or mechanically treated waste (0.5 % TS), while the leaching of ammonium nitrogen was reduced (79 g/t TS) due to stabilisation during the aerobic phase of treatment. The content of organic materials (1-4 g/l COD) and concentration of ammonium nitrogen (100-400 mg/l) in leachate during the initial phases (first 2 years) of mechanically-biologically treated MSW residuals was mainly lower than in the leachate of the 50-year-old MSW landfill (mainly 1-10 g/l COD and 300-2100 mg/l NH_4-N) in methanogenic phase.

TABLE 10 Landfill characterization methods used in the present study.

Characterization method	Aim of characterization	Sample size	Sample processing	Parameters	Vertical variation	Horizontal variation	Temporal variation
Landfill waste sampling	To study unknown content	Preferably >10 l up to hundreds of litres	Particle size from <2 mm to 50 mm for analyses and testing	TS, VS, pH, BMP, TKN, leaching of NH_4-N and COD	TS, VS, pH, BMP, TKN	All parameters	Monitoring not feasible due to slow progress of degradation
Internal leachate sampling	Conditions and stages of degradation	From a few deciliters to a few litres depending on parameters studied	Filtration for determination of soluble parameters	COD, BOD, VFA, pH, Redox, NH_4-N, TKN, Conducitivity, Cl	All parameters	All parameters	All parameters
On line monitoring	Water table, temperature and leachate quality	--	Not needed	Water table, temperature, conductivity, pH	Temperature, conductivity, pH	Water table, temperature, conductivity, pH	Possible, especially if high amounts of precipitation or leachate recirculation
Pore gas	Gas generation, stages of degradation	--	Not needed	CH_4, CO_2, O_2	CH_4, CO_2, O_2	CH_4, CO_2, O_2	CH_4, CO_2, O_2
Gas emissions	Gas generation and emissions, monitoring of methane oxidation besides pore gas concentrations	--	Not needed	CH_4, CO_2, N_2O	--	CH_4, CO_2, N_2O	CH_4, CO_2, N_2O
Gas collection	Gas generation, recovery efficiency	--	Not needed	CH_4, CO_2, O_2	Possible	Exist	Possible

TABLE 11 Characterisation of four different kind of landfills.

Landfill type/age	Waste management and landfill history	Structure	Organic material (mean % TS)	Methane potential (m^3/t TS)	TKN (mean % TS)	SCOD leaching (mean kg/t TS)	TKN leaching (g/t TS)
50-year-old MSW landfill	Incineration residuals from 1960s to 1980s, leachate recirculation, sludges, surplus soils, source segregation started in 1990s	Highly stratified, inert material at the bottom, may have high water table	40 Lower content below water table	6-60, mean 34, well stabilized in bottom layer <10 m^3/t TS	0.4 Vertical trend, lowest content in bottom layer	6[1] Lowest leachability in bottom layer	390[1] Lowest leachability in top layer
20-year-old MSW landfill	Non segregated MSW, source segregation started in 1990s	Highly heterogenous, high water table possible	63 High content in all layers	1-183, mean 50, high variation	0.4 Vertical trend, lowest content at bottom layer	19[1] Lowest leachability in bottom layer	841[1] Lowest leachability in top layer
Mechanically treated MSW residuals, newly established landfill	Source segregation and mechanical processing	Well mixed, small particle size, metals, high calorific waste segregated, density >0.9 t/m^3	61 Similar than in 20-year-old MSW landfill	259 High potential	0.5 Similar to that in old MSW landfills	24[2] Enhanced leachability compared to old MSW landfills	391[2] Enhanced leachability compared to old MSW landfills
Mechanically-biologically treated MSW residuals, newly established landfill	Source segregation and mechanical-biological processing	More homogenous than M residuals, density >1.0 t/m^3	43 Similar than in 50-year-old MSW landfill	52 Approximately 80 % reduced owing to aerobic treatment	0.6 Similar to that in old MSW landfills and mechanically treated residuals	0.6[2] 98 % reduced leachability compared to mechanically treated residuals	79[2] 80 % reduced leachability compared to mechanically treated residuals

[1] Leaching test, [2] During 426 days in landfill lysimeter

6 CONCLUSIONS

Waste and leachate sampling and on-line monitoring are important methods in seeking to characterise waste composition, landfill conditions (e.g., water content, temperature and pH) and changes in those conditions, and stages of degradation or stabilisation in old MSW landfills with unknown content. In new landfills with known content, the organic matter and nitrogen budget in addition to leachate and landfill gas characterisation are also feasible in order to characterising landfills during landfilling. Determination of the organic matter and nitrogen budget may provide a more advanced characterisation, as the initial contents (e.g, organic matter, methane potential, nitrogen) are known and the remaining contents can be estimated by the amounts of leached or volumes of landfill gas produced. In general characterisations of this kind can provide information for both active landfill and post-landfill operations in both old and new landfills.

To characterize MSW landfills requires intensive internal sampling and preferably large waste samples (up to hundreds of liters). The data obtained seem to be case-dependent especially in old MSW landfills with unknown content. It is important that sampling systematically covers both vertical and horizontal variation in landfills several decades old. The minimum number of sampling points will vary according to the parameters studied, the properties of the landfill and the accuracy required – thus general recommendations on the minimum number of sampling points cannot be given. However, in landfills approximately 30 m deep from 3 to 4 sampling depths would be needed at each sampling point to characterize the vertical layers.

On-line monitoring of the water table, temperature, electrical conductivity and pH of the internal leachate is feasible in MSW landfills. The present results also showed that the height of the water table affects internal leachate quality; thus it is important to consider the variation in the water table between samplings. However, seasonal variation was not seen in the water table or temperature in the internal leachate, even where the ambient temperature varied from -28 ^{o}C to 30 ^{o}C. Thus the utility of on-line monitoring in traditionally operated landfills and landfills in which a small amount (range 300-600 mm) of leachate

is recirculated is case-specific, as changes in the pH and conductivity of the internal leachate were also typically slow.

The 50- and 20-year-old MSW landfills contained varying amounts of biodegradable materials, as shown by the fact that the content of organic materials (VS/TS %) varied in samples from 3 to 80 % and from 35 to 89 %, and BMP from 1 to 183 m^3/t TS and from 6 to 60 m^3/t TS in the older and younger landfills, respectively. Moreover the NH_4-N leaching (range 0-2205 g/t TS and 0-1400 g/t TS) and COD leaching (2.6-51.4 kg/t TS and 0.5-15.9 kg/t TS) was higher – in some cases even several fold - in the 20-year-old than 50-year-old landfill, while TS was lower in the 20- year-old (range 35-81 %) than 50-year-old landfill (range 56-89 %). Nitrogen contents, however, was similar (mean approximately 4 g/kg TS) in both landfills. Thus it appears that landfills several decades old have high vertical and horizontal variability in waste quality, indicating that both the waste composition and state of degradation varied greatly in the 50- and 20-year-old landfills. Stabilization, especially with regarding to the content of organic materials and BMP, varied more vertically than horizontally, thus showing the effect of a prolonged period of stabilization as the vertical trend in BMP and organic materials rather than variation is caused by other factors. In most of the studied parameters marked differences indicated that local waste management and landfilling practices between landfills were different, even if both landfills had been receiving waste from the similar types of communities and were located in a similar climatic zone. The both 50- and 20-year-old landfills also had some characteristics in common, such as the highest nitrogen content (means 4.0 – 5.2 kg/t TS and 4.6 kg/t TS) and BMPs (means 44 m^3/t TS and 68 m^3/t TS) occurring in middle and top layers, while the bottom layer was the most stabilised, especially with regard to BMPs (means 8 m^3/t TS and 21 m^3/t TS).

The internal leachate in the 50-year-old landfill was typical for the methanogenic phase of degradation (pH 6.9-8.7, BOD/COD ratio mainly below 0.15), even if the higher vertical than horizontal variation indicated varying stages of degradation according to the vertical profile of the landfill body. The leachate in the upper part of the water saturated zone in the landfill indicated the presence of higher amounts of transportable organic material (COD, range mainly 4000-8000 mg/l) and nitrogen (NH_4-N, range mainly 1000-2500 mg/l) than in the lower parts (COD range mainly 500-2000 mg/l and NH_4-N range 300-1000 mg/l). The vertical variation can on the one hand partly be explained by the fact that the material in the 50-year-old landfill was more stabilized in the bottom layers, while on the other hand dilution stream in the bottom layers appears to proceed either horizontally from more stabilized areas or along preferential pathways through the landfill, or it may be owing to groundwater due to the absence of bottom structures. Additionally waste handling and landfilling practices were different in the bottom layers than in the top layers.

The determination of biological methane generation potential by means of carefully planned sampling appears to be a feasible method of determining first order kinetic factors (rate k and potential L_o) for bulk waste. Considerably different rates of methane generation (k of 0.05 and 0.17 in the 50- and 20-year-old landfills) and methane generation potential (L_o of 40 m^3/t and 130 m^3/t)

landfills) and methane generation potential (L_o of 40 m^3/t and 130 m^3/t) were demonstrated between the landfills. The different methane generation rates were also seen in the fact that actual methane generation rate (per ton of waste) was up to 15-fold in the 20-year-old than 50-year-old landfill. It was also found that high (41 % and 19 % of total potential) methane emissions before the installation of gas recovery systems may also occur in landfills located in boreal conditions, and that emissions into the atmosphere may account up to 60 % of the total methane generation potential, even if gas recovery is implemented several years before landfill closure.

Mechanically and mechanically-biologically treated MSW residuals can be characterized by their nitrogen and organic matter budgets in addition to their leachate and gas properties during landfilling as the initial contents are known. It seems that mechanically treated MSW residuals promote the acidogenic phase of degradation (pH 5.8-6.1) by at least two years as well as high rate leaching of ammonium nitrogen (range 600-1800 mg/l; load 391 g/t TS, 8 % of initial nitrogen content) and organic matter (range 20-100 g/l; load 24 kg COD /t TS). In contrast, the methanogenic phase of degradation (pH >7, BOD/COD ratio <0.2) was reached after a few months landfilling in mechanically-biologically treated waste, and the leaching of ammonium nitrogen (range 100-400 mg/l; load 79 g/t TS, 1 % of initial content) and organic matter (1-4 g/l; load 0.6 kg/t TS) was significantly lower during the 14 month study period.

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YHTEENVETO (Résumé in Finnish)

Jätteiden loppusijoittaminen kaatopaikoille on eri puolilla maailmaa edelleenkin yleinen yhdyskuntajätteiden käsittelymenetelmä. Nykyisin esimerkiksi EU-lainsäädäntö ja kansalliset päätökset ohjaavat jätehuoltoa ja jätteiden loppusijoitusta muihin käsittelymenetelmiin. Jätehuollon tavoitteena on lisätä materiaalien kierrätystä ja jätteiden energiasisällön hyödyntämistä energiantuotannossa sekä vähentää erityisesti biohajoavien jätteiden loppusijoittamista kaatopaikoille, sillä yhdyskuntajätteiden kaatopaikat ovat merkittäviä kasvihuonekaasu- ja vesistöpäästöjen aiheuttajia. Toisaalta kaatopaikalle loppusijoitettujen jätteiden metaanienergiapotentiaalia voidaan hyödyntää energiantuotannossa. Kaatopaikkojen jälkihoitovaiheen operointi (kaasunkeräys ja hyödyntäminen, suotovesien käsittely, rakenteiden toiminnan ja jätteiden hajoamisprosessien seuranta) kestää vuosikymmeniä tai jopa vuosisatoja jätteiden ja jätetäytön ominaisuuksista, ilmasto-olosuhteista sekä tavoitetasosta riippuen. Vanhojen ja suljettavien kaatopaikkojen jätetäytöt ovat heterogeenisia ja niiden koostumus ja ominaisuudet kuten metaanintuottopotentiaalit ja typpisisältö ovat usein huonosti tunnettuja. Tämä vaikeuttaa jälkihoitovaiheen operoinnin suunnittelua, energiapotentiaalin hyödyntämistä ja päästöjen käsittelytarpeen arviointia. Tässä tutkimuksessa tarkasteltiin erityyppisten kaatopaikkojen ominaisuuksien karakterisointia jäte- ja vesinäytteillä, jatkuvatoimisin sisäisen veden (taso, lämpötila, johtokyky, pH) mittauksilla, huokoskaasu- (CH_4, CO_2 ja O_2) ja päästömittauksilla (CH_4, CO_2 ja N_2O) sekä metaanintuoton mallinnuksilla. Tavoitteena oli kehittää menetelmiä kaatopaikkojen täyttö- ja jälkihoitovaiheen hajoamistilan ja olosuhteiden monitorointiin, operointitoimenpiteiden kuten suotoveden kierrätyksen ja kaasunkeräyksen suunnitteluun sekä tutkia jätteiden hajoamista ja hajoamisolosuhteita pohjoisissa ilmasto-olosuhteissa. Kenttätutkimukset suoritettiin kahdella, n. 20 (Ämmässuo, Espoo) ja n. 50 (Kujala, Lahti) vuotta käytössä olleilla ja vuoden 2007 aikana käytöstä poistettavilla kaatopaikoilla (1-6 hehtaarin koealueilla). Lisäksi tutkittiin alkutilan ominaisuuksiltaan tunnettujen mekaanisesti ja mekaanis-biologisesti käsiteltyjen yhdyskuntajätteiden ominaisuuksia ja käyttäytymistä loppusijoituksen aikana. Tämä tutkimus osoitti, että vanhoja jätetäyttöjä voidaan karakterisoida jätteiden ja sisäisen veden omaisuuksien perusteella. Karakterisoinnin perusteella jätetäytöt ovat kuitenkin hyvin heterogeenisiä ja voivat erota ominaisuuksiltaan huomattavastikin toisistaan. Siksi yksittäisen kaatopaikan karakterisoinnin tuloksia ei voida yksiselitteisesti soveltaa toisten kohteiden karakterisointiin. Tutkimuksessa lähes kaikki jätetäyttöjen näytteistä ja sisäisestä vedestä määritetyt parametrit osoittivat suurempaa vertikaalista kuin horisontaalista riippuvuutta jätetäyttöjen ominaisuuksissa, mikä osaltaan osoitti jätteiden stabiloituneen enemmän jätetäyttöjen alaosissa, missä suurempi kosteus ja veden liike todennäköisesti tehostavat hajoamista. Myös labotoriokokeissa kosteuden lisäys tehosti merkitsevästi jätteiden hajoamista. Jätenäytteiden keskimääräiset metaanintuottopotentiaalit ja typpipitoisuudet olivat suurimmat jätetäyttöjen keski- ja yläosissa (44-

68 m^3 CH_4/t TS ja 4-5.2 kg TKN/t TS TS) ja pienimmät jätetäyttöjen alaosissa (8-21 m^3 CH_4/t TS ja 2.4 kg TKN/t TS). Molemmilla tutkituilla vanhoilla kaatopaikoilla huomattava osuus metaanintuottopotentiaalista oli vapautunut päästöinä ilmakehään ennen kaasunkeräysjärjestelmän käyttöönottoa. Suotoveden kierrätyksen ei todettu yleisesti vaikuttavan sisäisen veden lämpötilaan tai ominaisuuksiin, vaikka yksittäisissä mittauspisteissä viitteitä vaikutuksista oli havaittavissa. Jatkuvatoimiset sisäisen vesipinnan, lämpötilan, pH:n ja johtokyvyn mittaukset toimivat pääsääntöisesti luotettavasti, ja ne osoittivat, että muutokset vanhojen kaatopaikkojen sisäisen veden laadussa ovat hitaita. Näin ollen jatkuvatoimisten mittausten käyttökelpoisuus ja tarpeellisuus edellyttää tapauskohtaista tarkastelua. Laitosmaisesti esikäsitellyt jätteet kuten mekaanisesti ja mekaanis-biologisesti käsitellyt jätteet ovat tasalaatuisempia kuin esikäsittelemättömät jätteet, mikä edesauttaa niiden ominaisuuksien karakterisointia. Tasalaatuisuus mahdollistaa niiden päästöpotentiaalin karakterisoinnin ja seurannan osittain myös massatasein. Esimerkiksi mekaanis-biologisesti käsitellyn jätteen typpitasetta voidaan seurata virtaamamittauksin, näytteenotoin ja analyysein, kun alkutilan typpipitoisuus ennen loppusijoittamista kaatopaikalle tunnetaan. Mekaanis-biologisesti käsitellyn yhdyskuntajätteen metaanintuottopotentiaali (52 m^3/t TS) oli n. 20 % mekaanisesti käsitellyn jätteen metaanintuottopotentiaalista (259 m^3/t TS). Molempien jätteiden typpipitoisuudet (5 kg/t TS ja 6 kg/t TS olivat) olivat lähes yhtä suuret, kun taas orgaanisen aineksen määrä oli mekaanis-biologisesti käsittellyssä jätteessä (43 % kuiva-aineesta) alhaisempi kuin mekaanisesti käsitellyssä jätteessä (61 % kuiva-aineesta). Sen sijaan mekaanis-biologisesti käsitellyn jätteen suotoveden COD kuormitus (0.6 kg/t TS) oli vain n. 2 % mekaanisesti käsitellyn jätteen kuormituksesta (24.2 kg/t TS) ja mekaanis-biologisesti käsitellyn jätteen typpikuormitus (103 g/t TS) vastaavasti n. 15 % mekaanisesti käsitellyn jätteen typpikuormituksesta (694 g/t TS) n. 14 kk aikana.

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ORIGINAL PAPERS

I

DETAILED INTERNAL CHARACTERISATION OF TWO FINNISH LANDFILLS BY WASTE SAMPLING

by

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Detailed internal characterisation of two Finnish landfills by waste sampling

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Abstract

The aim of this study was to characterise the internal structure and composition of landfilled waste at two Finnish landfills to provide information for active and post-landfill operations. The two sites, Ämmässuo and Kujala, have been in operation for 17 and 48 years, respectively. Waste was sampled (total 68 samples) and analysed for total solids (TS), volatile solids (VS), total Kjeldahl nitrogen (TKN), biological methane potential (BMP) and leaching of organic material (determined as chemical oxygen demand, COD) and ammonium nitrogen (NH_4–N). The results showed high vertical and horizontal variability, which indicated that both the waste composition and state of degradation varied greatly in both landfills. Ämmässuo was characterised by 2- to 4-fold higher BMP, NH_4–N and COD leaching than Kujala. Moreover, the ratio of VS to TS was higher at Ämmässuo, while TS content was lower. The highest mean BMPs (68 and 44 m^3/t TS), TKN content (4.6 and 5.2 kg/t dry weight) and VS/TS ratio (65% and 59%) were observed in the middle and top layers; and the lowest mean BMP (21 and 8 m^3/t TS), TKN content (2.4 kg/t dry weight, in both landfills) and VS/TS ratio (55% and 16% in Ämmässuo and Kujala, respectively) in the bottom layers. In conclusion, waste sampling is a feasible way of characterising the landfill body, despite the high variation observed and the fact that the minimum number and size of samples cannot easily be generalized to other landfills due to different methods of waste management and different landfilling histories.

1. Introduction

Landfilling has continued to be a major method for municipal solid waste (MSW) disposal during recent years (Eurostat, 2005; US EPA, 2005a). In the European Union (EU), solid waste management and landfilling are undergoing major changes following the EC Landfill Directive (1999/31/EC). The requirement that landfills must have a bottom liner means that a large number of landfills in the EU will be closed by 2007 (EC, 1999). The EU directive also phases out the quantity of organic waste that can be landfilled; therefore, waste minimisation and pre-treatment before landfilling are encouraged, which in turn affects the composition of landfilled waste. From the 1990s, Finnish MSW has been increasingly segregated at source into biowaste, glass, metals, paper and cardboard and residual fraction. The residual fraction as such or after mechanical removal of materials for recycled fuel or other uses, the residual fraction has commonly been landfilled.

Operational and closed landfills are potential sources of environmental pollution, such as polluted leachates (e.g., ammonia and dissolved constituents; Ehrig, 1989) and greenhouse gases (e.g., methane; IPCC, 2001). When landfills are closed, they are typically sealed by cover layers such as geomembrane composites and/or soils to minimise leachate generation and gas formation or emissions. The landfill gas can be collected and used for energy production, flared, or alternatively methane can be oxidised biologically into carbon dioxide. It has been estimated that waste degradation and emissions from waste in landfills will continue for decades or even centuries after closure (e.g., Stegmann, 1989). However landfills can also be operated as bioreactors in order to enhance biodegradation and

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stabilisation with a higher level of control for liquids and gases. A typical bioreactor landfill operation would apply leachate recirculation to adjust the moisture and improve the contact between different substrates and micro-organisms, and thus stimulate anaerobic degradation (e.g., Reinhart and Al Youshi, 1996; Reinhart and Townsend, 1998; Morris et al., 2003). Recently, landfill aeration to enhance biodegradation of waste has also been studied (Heyer et al., 2005).

Because in many cases the composition, volume and placement of landfilled wastes were not well documented, the internal composition and structure of landfilled wastes and their emission potential remain unknown. In the landfill, wastes undergo various biological, physical, and chemical processes at different rates. These processes, together with the heterogeneous nature of the landfilled waste, may result in different conditions in different parts of the landfill. All of these factors can cause uncertainty when planning post-landfill and/or bioreactor operations. In practice, the content of the landfill is mainly dependent on the specific waste management practices of communities contributing waste to a particular site. The major fractions (Table 1) of discarded MSW are paper and cardboard, kitchen biowaste, plastics and garden waste (Golder Associates, 1999; YTV, 2004; US EPA, 2005b), and major portions of the methane potential can be attributed to cellulose and hemicellulose (Barlaz et al., 1989; Baldwin et al., 1998). The emission (and energy) potential of different MSW fractions vary greatly; e.g., the source segregated residual fraction of MSW (termed "grey waste" in Finland) and biowaste may have a biological methane potential (BMP) of 46 m^3/t total solids (TS) (grey waste) and 410 m^3/t TS (biowaste) and contain 2.1 kg NH_4–N/t TS (grey waste) and 3.6 kg NH_4–N/t TS (biowaste) of leachable nitrogen (Jokela et al., 2002). Furthermore, in addition to waste, landfills often contain soil of variable properties, which is used as daily cover.

Table 1
Composition of MSW (wet weight) after source segregation of recyclable materials in Finland, US and Australia

Waste	Finland (%)[a]	US (%)[b]	Australia (%)[c]
Paper and paper/cardboard	20	26.3	9.9
Glass	4	6.2	6.8
Metals	4	7.3	7.1
Plastics	13.6	15.4	7.3
Rubber and leather	–	3.5	–
Textiles	4	5.5	–
Kitchen biowaste	38 (including garden waste)	16.4	38.1
Garden waste	–	7.6	17.8
Wood	3	7.5	6.4
Inorganic Wastes	–	2.2	–
Diapers	7	–	–
Other combustible	3	–	–
Other	3.4	2	6.6

[a] YTV (2004).
[b] US EPA (2005b).
[c] Golder Associates (1999).

In order to evaluate the range of conditions prevailing in existing landfills, this study examined gas, pore water and leachate quality at two Finnish landfills. Due to the large size and heterogeneity of most landfills, and for economic reasons, internal landfill sampling must be carefully coordinated with available information about waste age and composition. Sampling before or in conjunction with the construction of gas recovery wells or leachate recirculation, especially at sites with limited existing information, may have economic benefits to optimise the gas extraction or liquid recirculation methods. Landfills have been previously sampled to estimate the rate of degradation of MSW and its different waste components (e.g., Hartz and Ham, 1983; Bogner, 1990; Gurijala and Suflita, 1993; Baldwin et al., 1998; Jokela et al., 2002; Gardner et al., 2003) while – to our knowledge – only a few studies have been published on vertical profiles of pH, temperature, moisture, organics, cellulose, lignin, or BMP (Bookter and Ham, 1982; Jones et al., 1983; Attal et al., 1992; Ham et al., 1993; Wang et al., 1994; Townsend et al., 1996; Chen et al., 2004; Östman et al., 2006) and even fewer studies (Ettala et al., 1988; Ham et al., 1993; Östman et al., 2006) on landfill nitrogen content. These earlier studies showed MSW landfills to be heterogeneous with respect to the stages of degradation and conditions within the landfill body with wastes in the top layers usually less degraded than in the deeper layers.

The aim of this study was to evaluate the feasibility of sampling landfill bodies (and analysing the samples) in order to characterise their properties and thus provide information for post-landfill monitoring and operation. The specific objectives were (1) to analytically characterise the composition, TS, volatile solids (VS), total Kjeldahl nitrogen (TKN), pH, BMP and NH_4–N as well as organic material (determined as chemical oxygen demand, COD) leaching in two Finnish MSW landfills; and (2) to characterise their vertical and horizontal distribution.

2. Materials and methods

2.1. Sampling sites and sampling

The samples were taken from the MSW landfills of Ämmässuo (Espoo, Finland, 17 yr in operation) and Kujala (Lahti, Finland, 48 yr in operation) (Table 2). The sampling was done during the installations of vertical gas collection wells (borehole ∅ 1.2 m, Ämmässuo) or leachate observation tubes (borehole ∅ 90 mm, Kujala). The sampling procedures were planned to obtain depth profiles at different locations using a 50–100 m grid (Fig. 1). For both landfills the mean values and standard deviations of the measured variables were calculated for different layers, which were normalized according to age (Table 3) using, as far as possible, a statistically significant number of samples from each layer (Table 4).

In the Ämmässuo landfill (the largest landfill in Scandinavia, established in 1987 in metropolitan Helsinki), the

Table 2
Characterisation of Ämmässuo and Kujala landfills

Parameter	Ämmässuo landfill	Kujala landfill
Established in	1987	1955
Landfilled waste (1000 t/a)	500–600	20–70
Area (ha)	54	27
Waste amount (million t wet)	7.4 (in 2004)	3.2 (in 2003)
Altitude (m from sea level)		
Bottom structure	+60–62	+99–100
Max. altitude (at sampling points)	+94 in 2004	+123 in 2003
Water table	+68–72 in 2003	+104–110 in 2003
Start of source segregation		
Biowaste	1995[a]	1994[a]
Energy waste		1998[a]
Paper and cardboard	During 1990s[b]	1998[a]
Other (e.g., glass and metals)	Area collection 1990s	Area collection 1990s
Annual rainfall (mm)	601–700[c]	601–700[c]
Leachate recirculation or irrigation	Not used in sampling area	Irrigation in sampling area in 1970–2000
Leachate formation (m^3/a)	85,775 (in 2000)[d]	55,785 (in 2003)[e]
Leachate characteristics[d]		
NH_4–N (mg/l)	570	43–210 (mean 112)
N_{tot} (mg/l)	580	62–222 (mean 126)
COD_{cr} (mg/l)	1600	250–550 (mean 367)
TOC (mg/l)	–	110–170 (mean 140)
BOD_7 (mg/l)	210	80[d] (one analysis)
pH (in observation tubes, own data, not shown)	6.0–7.5	6.8–8.2

[a] Residential buildings containing more than 10 apartments.
[b] Residential buildings containing more than 20 apartments.
[c] Finnish Meteorological Institute, 2006.
[d] Annual mean values and/or ranges measured from regular monitoring of leachate quality (in Ämmässuo: Soil and Water Ltd., 2002; and in Kujala: Vääränen and Tuominen, 2004; Vääränen, 2005).

total amount of landfilled waste is about 8 million tons. Waste is currently being deposited at an annual rate of 0.5 million tons, of which 0.3–0.4 million tons is MSW and the rest is mainly industrial and construction waste (Table 2; Karhu, 2004). Initially, all of the mixed MSW was landfilled; however, during the 1990s source segregation (biowaste, paper and cardboard, glass and metals) gradually increased in this region and from the end of that decade, only the residual fraction of MSW (grey waste) has been landfilled. Historically, MSW has been disposed in three sectors of this site (sectors referred as 1, 2 and 3, Fig. 1) so that a similar waste history can be assumed across the landfill. Waste was compacted in horizontal layers with soils, composted sewage sludge or biowaste as daily cover. For this study, samples from sector 3 (40 samples) and sector 1 (4 samples) were taken: in sector 3 from

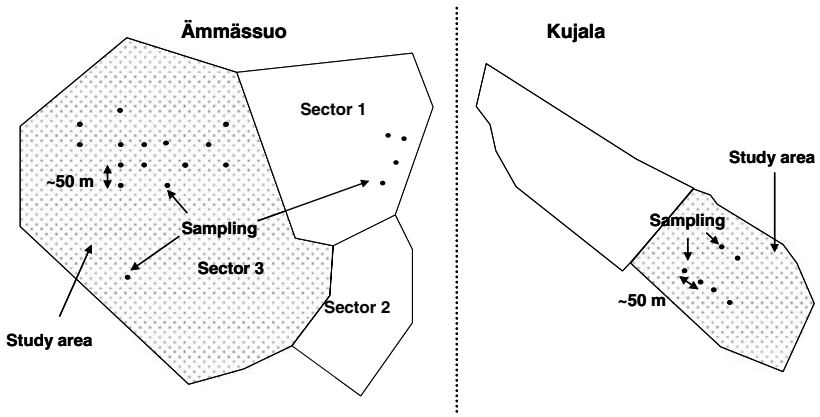

Fig. 1. Sketch of locations of sampling points at Ämmässuo and Kujala landfills.

Table 3
Approximate years of landfilling type of wastes at depths sampled in this study (based on information from the two sites)

Level (m)	Years of landfilling	Landfilled waste
Ämmässuo landfill		
80–88	1998–2003	Grey waste
72–78	1993–1996	MSW, some biowaste source segregated
62–66	1987–1992	MSW
Kujala landfill		
118–121	1998–2003	Grey waste
109–114	1995–1997	MSW, some biowaste source segregated
106–108	1984–1989	Surplus soils, MSW
99–103	1960–1974	Surplus soils, ashes, assorted wastes, sewage sludge

two to four depths at 15 boreholes (∅1.2 m) and in sector 1 at 4 boreholes (one depth) 1 by grab sampling during the installation of the gas collection wells using a drilling rig. Waste materials were crushed during drilling so that the largest particles were typically <20 cm in diameter. Larger fragments of metals, plastics, and stones, which were not crushed, were removed and weighed during sampling. The weight of all removed materials was included in the results. The samples were taken from +62 to +82 m above mean sea level; the bottom of the waste was +60 to +62 m above mean sea level. The temporary cover of the landfill varied from +82 to +94 m at the sampling points; hence the sampling levels were reported from mean sea level. The volume of each sample was about 300–500 l. Samples were spread out on a concrete floor in layers of 20–40 cm from which two replicate randomised composite samples (about 30 l) were shovelled into plastic bags, one for sorting/description and the other for analyses. The sorting was done within 2 days, while the analytical sample was stored for 1–2 months at ambient temperatures (below 0 °C) prior to analysis.

The Kujala landfill (Lahti, Finland) was initially established for surplus soils and MSW (Ettala et al., 1988). The amount of landfilled waste was approx. 3.2 million tons in 2003. From 1965 to 1984, the landfilled material consisted mainly of incineration residuals of MSW. In 1965, approx. 200 tons of MSW and during 1975–1984 over 20,000 tons of MSW were incinerated. Sewage sludges were landfilled until 1980. Thereafter the rate of landfilling was increased from 20,000 (in 1981) to 67,000 tons (in 2004), of which about 44,000 tons was classified as MSW while the rest was industrial (ash, wood dust) and construction waste (Table 2; Päijät-Häme Waste Management Ltd., 2005). MSW was landfilled from 1984 until 1994 when biowaste source segregation was implemented. The landfilled waste underwent further changes after 1998 when source segregation of MSW into biowaste, energy waste, paper, cardboard, metal, glass and landfill waste was introduced. Waste is landfilled in horizontal layers and compacted by a landfill compactor using soil for daily cover. Waste was commonly burnt on Finnish landfill sites from the 1960s until the 1980s; in addition, leachate recirculation via sprinklers was practiced during the summer from the 1970s to the 1990s. The bottom level of the landfill (no liner) is at +99 to +100 m above mean sea level and the temporary cover layer was at +115 to +23 m above mean sea level at the time of this study (in 2003). The waste in the study area was landfilled from the 1960s to 2003 (Table 3). The waste samples (24) from six boreholes and four sampling levels were taken during the installation of observation tubes by a drilling rig from level +101 m to +122 m. This drilling procedure crushed most samples into particles of <50 mm diameter. The samples obtained were representative of about 1 m of the depth profile. The samples (10–20 l size) were stored in airproof plastic bags outdoors (below 0 °C) for 2–3 months before sorting and analyses. Some bulk materials (mainly stones, metals

Table 4
Mean characteristics (± standard deviations) of samples at various depths

Sample level (m)	TS (%)	VS_{wet} (%)	VS/TS (%)	TKN (kg/t dry weight)	NH_4–N leaching (g/t TS)	NH_4–N leaching/TKN ratio (%)	COD	BMP leaching (kg/t TS)	pH (m^3/t TS)
Ämmässuo landfill									
80–88	57 ± 10	35 ± 7	64 ± 14	4.6 ± 2.8	625 ± 655	13 ± 14	22 ± 14	47 ± 55	7.0
72–78	52 ± 11	33 ± 9	65 ± 15	3.5 ± 2.1	1120 ± 630	37 ± 28	20 ± 8	68 ± 61	7.4
62–66	50 ± 10	27 ± 1	55 ± 11	2.4 ± 1.4	850 ± 511	35 ± 22	12 ± 5	21 ± 25	7.6
Mean ± stdev	54 ± 10[1]	33 ± 8[1]	63 ± 14[1]	3.9 ± 1.5[1]	841 ± 648[2]	25 ± 23	19.3 ± 11.1[2]	50.1 ± 54.6[3]	7.2
Kujala landfill									
118–121	67 ± 3	35 ± 5	51 ± 3	4.0 ± 1.8	109 ± 201	3 ± 6	3.4 ± 1.9	44 ± 15	–
109–114	65 ± 5	35 ± 9	59 ± 17	5.2 ± 2.1	684 ± 481	11 ± 7	9.7 ± 5.8	21 ± 19	–
106–108	67 ± 4	24 ± 7	36 ± 11	4.6 ± 2.1	493 ± 368	12 ± 9	6.0 ± 3.1	22 ± 17	–
99–103	73 ± 11	12 ± 9	16 ± 8	2.4 ± 0.8	170 ± 126	7 ± 4	2.5 ± 3	8 ± 2	–
Mean ± stdev	68 ± 7[4]	26 ±12[4]	40 ± 21[4]	4 ±2[4]	390 ± 396[4]	9 ± 7	5.6 ± 4.8[4]	33.8 ± 35.4[5]	–

Number of samples analysed [1]40, [2]34, [3]32,[4]24, [5]11.

and glass), which are difficult to shred, were removed and the proportion of these materials was included in the sorting results, but excluded from the analyses.

2.2. Sorting and processing of samples

Waste samples were manually sorted into seven (Ämmässuo) or five (Kujala) categories, namely plastics, paper and cardboard, wood, metals, inert materials (e.g., glass, stones), textiles and residuals (Fig. 2). Textiles were included in the residuals and metals in the inert materials when sorting the Kujala samples. Sorting was based on visual inspection, and thus small particles, which could not be visually identified, were classified as "residuals". Each category was weighed separately.

For analytical purposes the samples were shredded (Retsch SM 2000) into three size fractions, except bulky materials which were not processed (e.g., metals and stones). The particle size of the processed fractions was <2 mm for TKN, <20 mm for the TS and VS analyses, and <50 mm for the methane production assays and shaking leaching tests. The bulky materials that were removed were weighed and taken into account in the sorting, but not in the analyses. This means that compared to the actual landfill conditions, some of the variables (e.g., TKN and BMP) in the samples had higher observed concentrations relative to their in situ condition, according to the proportion of materials removed.

2.3. Analyses and calculations

BMPs were determined with digested mesophilic municipal sewage sludge (Nenäinniemi Jyväskylä, Finland) as an inoculum (500–1300 ml) in duplicate 2-l glass vessels. The waste samples were added to the vessels to obtain the ratio of 2 g VS_{waste}/g $VS_{inoculum}$. According to our experience, this ratio usually enables methane generation without the accumulation of organic acids. The BMP of the inoculum was determined separately and was reduced from that of the samples and inoculum together. The final liquid volume of 1.5 l in each vessel was obtained by adding deionised water; thus the moistures in all the assays (with waste, inoculum, and water) were 88–95% of wet weight. The bottles were flushed with N_2/CO_2 (80/20%) and sealed with butyl rubber stoppers. The gas produced was collected in aluminium sampling bags (Tecobag PETP/AL/PE-12/12-75, Tesseraux Spezialverpackungen). The BMPs were continued until methane production became negligible (<5 ml CH_4 d^{-1}) after 70–100 d. In addition to BMP, the methane production of four samples from both landfills were determined with their original (32–57%) moisture content and at 60% moisture content with the water added in the batch assays without inoculum. The determinations were performed for the same four samples from Kujala as used in this study elsewhere, while for Ämmässuo four different samples (sampled from landfill sector 1) were used, and thus their BMP with inoculum was also determined. Three of the samples from both landfills were from the top layers (2–5 yr-old waste) while the fourth samples were from the

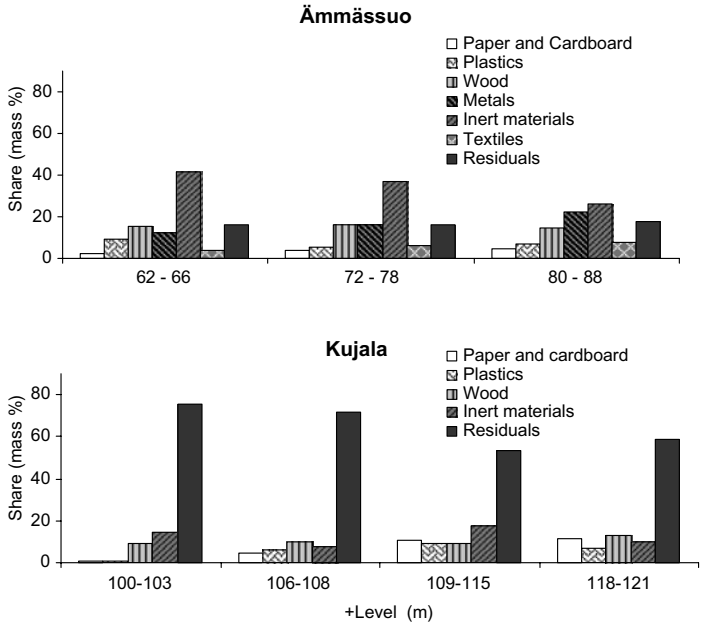

Fig. 2. Waste composition at different depths at Ämmässuo and Kujala landfills.

bottom layer (15- and 40 yr-old waste in Ämmässuo and Kujala, respectively). All methane production assays (including BMPs) were performed at 20–22°C.

TS and VS were analysed according to standard methods (APHA, 1998). Moisture content was determined by subtracting the analysed TS content from 100%. Leaching of NH_4–N and COD were determined by samples obtained in a one-stage shaking leaching test at a liquid-to-solid (*L*/*S*) ratio of 10 according to SFS-EN 12457-4 (2002), except that the samples were shredded below 50 mm, instead of the 10 mm particle size mentioned in the standard method. The samples for the NH_4–N and COD analyses were fil-

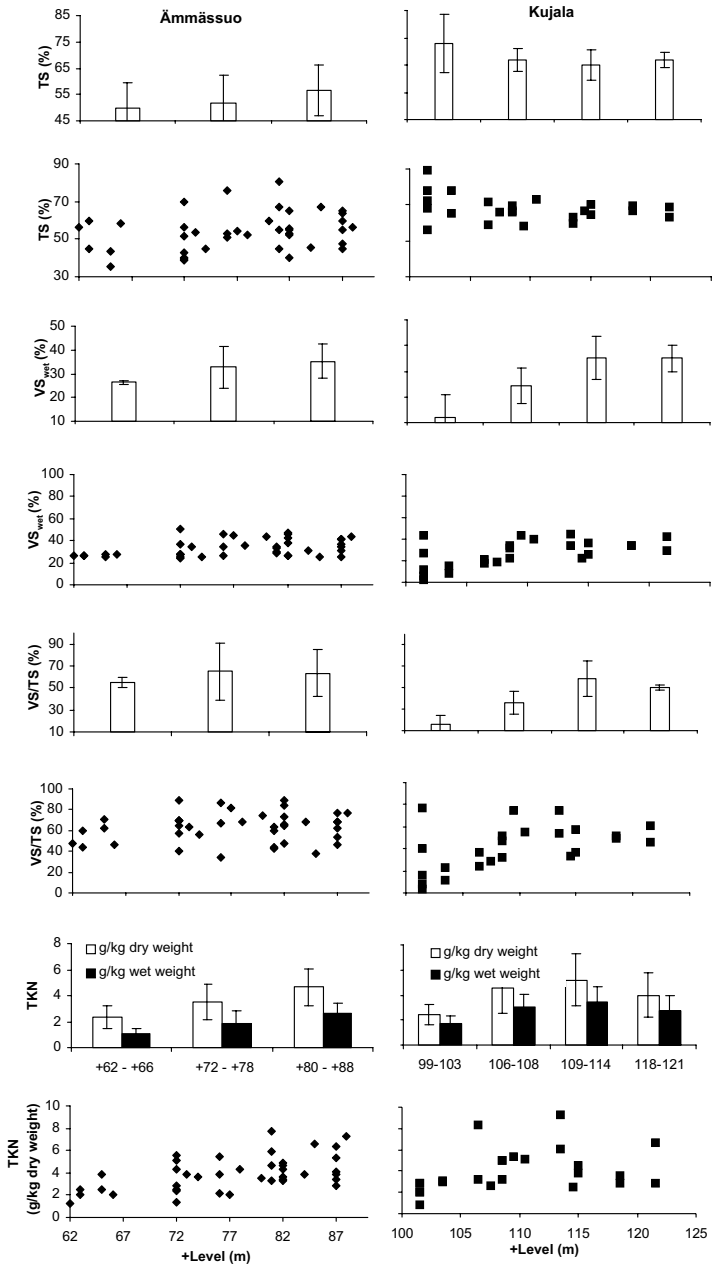

Fig. 3. TS, VS, VS/TS and TKN contents of waste samples with trendlines (and linear regression coefficients, if significant) along the depth profiles and mean (±stdev) values at different layers of Ämmässuo and Kujala landfills.

tered with GF50 glass fibre filter papers (Schleicher and Schuell). COD was analysed according to SFS 5504 (1998). TKN and NH_4–N were analysed according to the application note supplied by Tecator (Perstop Analytical/Tecator AB, 1995). A WPA CD70 pH meter and Sensorex pH 450 CD electrode were used to measure the pH of the Ämmässuo samples. Biogas volume was measured using a water displacement method in a special gas measurement cylinder. Methane content was measured by a Perkin–Elmer Autosystem XL gas chromatograph with a flame-ionization detector as described in Luostarinen and Rintala (2005).

3. Results

In terms of vertical and horizontal variability, clear trends appeared mainly with respect to sampling depth, although some different characteristics between individual samples were also observed horizontally (Figs. 1, 3 and 4). No clear spatial differences were detected (data not shown) horizontally across the landfills.

In Ämmässuo, the major weight fraction was inert materials (30–40%, mainly stones), while the other fractions contributed shares of <22% (Fig. 2). In Kujala, the residuals (soils and unrecognisable materials) formed the largest fraction at all depths (54 – 75%). The proportion of paper and cardboard was lowest in the bottom layer of both landfills (2% in Ämmässuo and 0.5% in Kujala), while the percentage of wood (15–16% in Ämmässuo and 9–13% in Kujala) was almost the same at all depths in both landfills.

The TS, VS, VS/TS ratio, pH (measured from the Ämmässuo samples) and TKN of the landfill body samples were studied to evaluate the properties and conditions within the landfills (Table 4 and Figs. 3 and 4). In Ämmässuo, pH varied from 6.7 to 8.9 and the mean pH at different layers increased from 7 to 7.6 towards the bottom of the landfill. In Kujala, the pH of the leachate (measured from observation tubes in the same location from which the samples were obtained) ranged from 6.8 to 8.2. The TS of individual samples ranged from 35% to 81% in Ämmässuo and from 56% to 89% in Kujala, while mean TS content was lower in Ämmässuo (54%) than Kujala (68%). Mean TS decreased (from top down) in Ämmässuo, while in Kujala no clear trend along the depth profile emerged. The mean VS/TS ratio of samples was 63% in Ämmässuo (range 35% to 89%) and 40% in Kujala (range 3–80%). In both landfills, the VS/TS ratio was lowest (mean 55% in Ämmässuo and 16% in Kujala) in the bottom layers. The TKN of individual samples ranged from 1.3 to 7.8 g/kg dry weight (mean 3.9 g/kg dry weight and 2.1 g/kg wet weight) and from 0.8 to 9.3 g/kg dry weight (mean 4 g/kg dry weight and 2.7 g/kg wet weight) in Ämmässuo and Kujala, respectively (Table 4 and Fig. 3). The mean TKN of the bottom layers were 52% and 60% (% dry weight) that of the top layers in Ämmässuo and Kujala, respectively.

The leaching of the NH_4–N and COD of the landfill body samples were studied by shaking leaching tests. NH_4–N-leaching was 0–2205 g/t TS (mean 841 g/t TS) in Ämmässuo and 0–1400 g/t TS (mean 390 g/t TS) in Kujala (Fig. 4). In Ämmässuo, the highest NH_4–N leaching was in the middle layer of the landfill and in Kujala the highest NH_4–N leaching was in the second (from top) layer. The NH_4–N-leaching/TKN ratio was 0–79% (mean 25%) in Ämmässuo and 0–24% (mean 9%) in Kujala. The highest NH_4–N leaching/TKN ratios were in the middle and bottom layer in Ämmässuo and in the two middle layers in Kujala, in spite of the fact that nitrogen content was lowest in the bottom layers of both landfills. COD leaching was about 4-fold higher in Ämmässuo (2.6–51.4 kg/t TS, mean 19.3 kg/t TS) than in Kujala (0.5–15.9 kg/t TS, mean 5.6 kg/t TS) (Table 4 and Fig. 4). In Ämmässuo, COD leaching decreased downwards in the landfill, while in Kujala the highest COD leaching was in the second (from top) layer.

The BMP ranged from 1 to 183 m^3/t TS (mean 50 m^3/t TS) in Ämmässuo and 6–60 m^3/t TS (mean 34 m^3/t TS) in Kujala (Table 4 and Fig. 4). In both landfills, the BMP was lowest in the bottom layer, while wide variation and the highest values (above 100 m^3/t TS in Ämmässuo and above 30 m^3/t TS in Kujala) were found in the upper layers.

The methane production of four samples from both landfills at their original moisture contents and at 60% moisture content started after a lag of 2–3 months (Table 5 and Fig. 5). Methane production at original moisture content was 0.0006–8.3 m^3/t TS (Ämmässuo) and 0.03–0.2 m^3/t TS (Kujala) after incubation for 544 (Ämmässuo) and 372 (Kujala) days. The adjustment of moisture content to 60% increased methane production to 1.6–63 m^3/t TS (Ämmässuo) and 0.06–13.4 m^3/t TS (Kujala), the highest increase for an individual sample being from 8.3 to 63 m^3/t TS for a sample with 50% original moisture. In the other words, methane production at the original moisture content was 0.0003–4.5% (Ämmässuo) and 0.2–1.0% (Kujala), while at 60% moisture content methane production was 2.3–34% and 0.9–32% of BMP in Ämmässuo and Kujala, respectively.

4. Discussion

The present study, which attempted to characterise landfill bodies approximately 20 and 40 yr old shows the difficulty attached to such an undertaking, but also provides some important information about the possibilities of characterising landfill contents and generalising the results to other landfills. Large variation in all the parameters studied was found in both landfills despite the fact that the samples were obtained by systematic vertical (3–4 heights) and horizontal (ca 50×50–100 m) sampling and that the initial sample size was relatively large (300–500 l in Ämmässuo, 10–20 l in Kujala) compared to the procedures used in some previous landfill studies (e.g., 0.5–1.0 l, Bookter and Ham, 1982; ca 100 l, Suflita et al., 1992; ca 1 l, Östman et al., 2006). It is evident therefore that characterisation of a heterogeneous landfill body requires

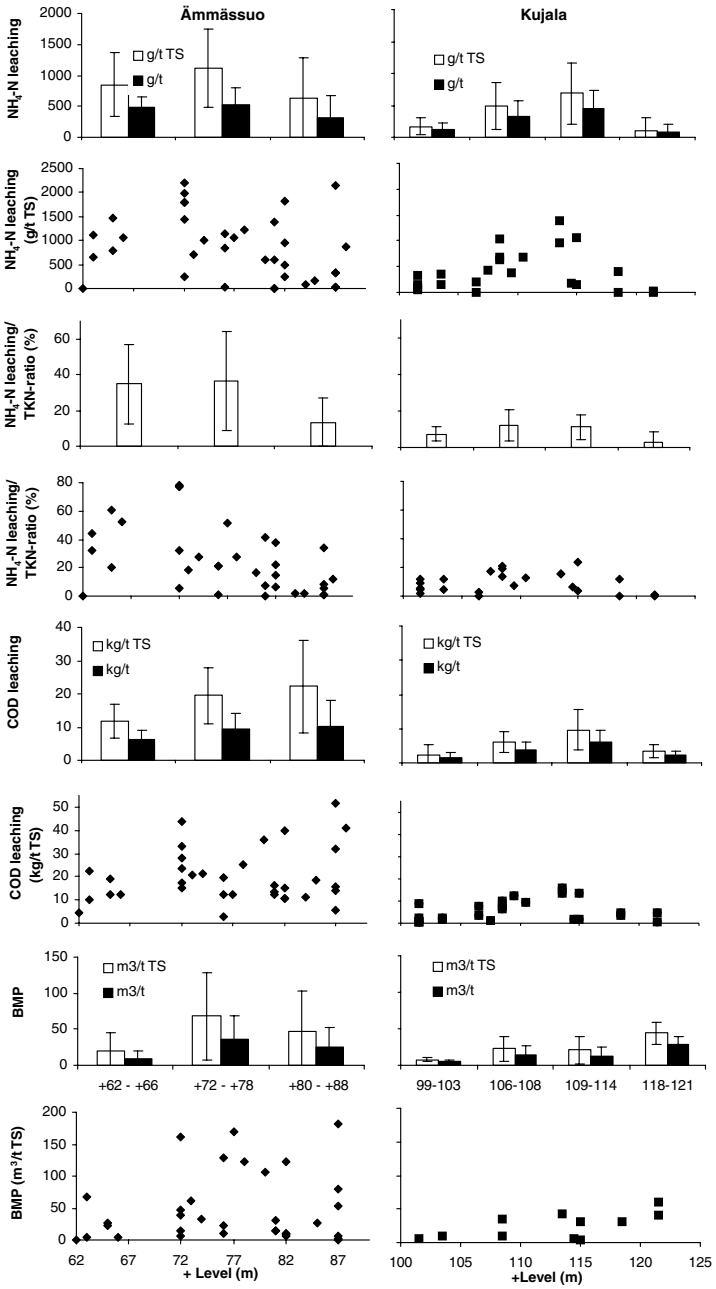

Fig. 4. NH_4–N leaching, NH_4–N leaching/TKN ratio, COD leaching and BMP of waste samples with trendlines (and linear regressions coefficients with equations, if significant) along the depth profile and mean values (±stdev) at different layers of Ämmässuo and Kujala landfills.

intensive sampling (Bookter and Ham, 1982; Suflita et al., 1992; Ham et al., 1993) and that the minimum number of sampling points varies according to parameters studied, landfill properties and the accuracy required – thus general recommendations on the minimum number of samples cannot be given. In the present study, extreme variation was observed: for example, in the highest 20 m layer in Ämmässuo landfill, where seven samples (of a total of 26 samples) had a BMP of 106–183 m^3/t TS, and eight samples had a BMP of less than 10 m^3/t TS. From 3 to 4 sampling depths would be needed at each sampling point in landfills 30 m deep to characterise vertical layers, as was done in this

Table 5
Methane production for samples at original and at 60% moisture contents and BMP of four samples from Ämmässuo and Kujala landfills

Sample level (m)	VS/TS (%)	Moisture content (%)	CH_4 production at initial moisture content (m^3/t TS)	CH_4 production at 60% moisture content (m^3/t TS)	BMP (m^3/t TS)
Ämmässuo landfill					
64	78	33	0.0006	8.5	182
73	65	35	0.02	nd*	193
69	69	50	8.3	63	183
63	66	57	0.8	1.6	70
Mean ± stdev	70 ± 6	44 ± 12	2.3 ± 4.0	24.4 ± 33.6	157.0 ± 58.2
Kujala landfill					
100	40	32	0.03	0.06	6.6
115	33	33	0.06	0.3	6.3
114	57	35	0.05	1.8	30
121	47	37	0.2	13.4	41.5
Mean ± stdev	44 ± 10	34 ± 2	0.09 ± 0.1	3.9 ± 6.4	21.2 ± 17.6

*Not determined.

study. At least one of the sampling depths should be below the water table where the waste is usually oldest and where the high moisture content and leachate movement favour stabilisation, as shown by Hartz and Ham (1983).

Both landfills had some common features, e.g., high proportions of soils or soil-like materials (including ash). These were apparently from daily soil cover, ash, and demolition waste. There were similar proportions of wood at both sites (15–16% in Ämmässuo and 9–13% in Kujala); other studies have indicated the recalcitrance of wood under landfill conditions (Micales and Skog, 1997; Gardner et al., 2003). In both landfills, the proportion of paper and cardboard decreased towards the bottom of the landfill, indicating their degradation. This is in accordance with the fact that their main components are cellulose and hemicellulose, which have been found to contribute up to 91% of the methane produced in landfills (Barlaz et al., 1989). Similar contents of plastics (5–9%) were found in all layers in both landfills, except in the bottom layer of Kujala, where its low content (1%) might be due to either low original percentages or on-site incineration at time of landfilling. The difference in inert contents between the two landfills (26–42% in Ämmässuo and 8–17% in Kujala) is probably smaller than found in the present sorting test,

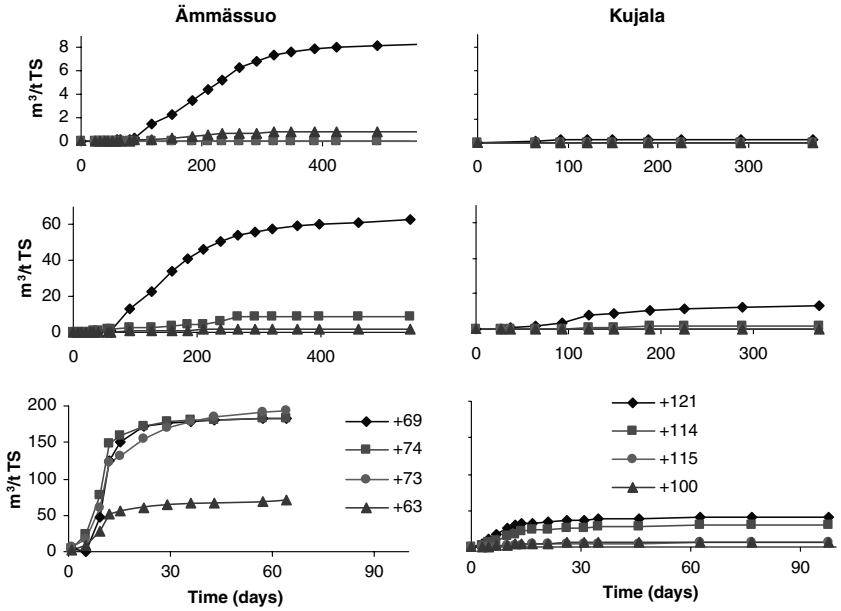

Fig. 5. Methane production from Ämmässuo and Kujala samples (named in figure by sampling depth above mean sea level) at original moisture content (up), at 60% moisture (without additional inoculum; middle) and in BMP assay (inoculum added; bottom).

because the drilling method used to sample Kujala excluded larger (>90 mm) waste particles (e.g., large stones, metals) from the samples. The proportion of residuals was smaller in Ämmässuo (16–18%) than in Kujala (54–75%), which was probably due to the increased proportion of residuals in Kujala where the sampling was done with a smaller borehole (90 mm) than Ämmässuo (1200 mm) and textiles were included among the residuals.

In general, the two landfills studied were very different, despite the facts that both were major MSW landfills in their regions, both received waste from similar communities, and both had similar climatic conditions in close proximity to each other (110 km). Thus, the straightforward application of information from one landfill to another one without sampling, as in this case from Kujala to Ämmässuo, is not feasible. In both landfills, some similar trends in vertical characteristics were observed. In general BMP, VS, VS/TS, NH_4–N and COD leaching were higher – some even several-fold – in Ämmässuo than in Kujala, while TS was lower in Ämmässuo than in Kujala. Furthermore, variation in all the parameters studied was clearly larger in Ämmässuo than in Kujala. These differences could indicate that waste, especially in the middle and top layers in Ämmässuo, contained significant amounts of biodegradable material which was probably just beginning to degrade. This type of material apparently also has high water holding capacity, thus decreasing the TS of the landfill. In Kujala, landfill practices (irrigation, on-site burning) and the landfilling of incineration ash have apparently resulted in a lower organic content in the bottom layers. In both landfills, the bottom layer differed from the upper layers in some variables. This was probably due to enhanced degradation and leaching below the water table, as the lowest samples from both landfills were taken from below water table, which was approximately 10 m from the bottom in Ämmässuo (height up to 34 m) and varied from 4 to 10 m in Kujala (height up to 24 m).

As discussed above, the VS/TS ratio in the two upper layers indicated a higher proportion of degradable material in Ämmässuo (VS/TS 64–65%) than in Kujala (51–59%), a difference which was even more significant in the bottom layer where VS/TS was 55% in Ämmässuo and 16% in Kujala. The lower VS/TS in the bottom layer was apparently due to biological stabilisation of the material over time; however, this was still incomplete even after some 20 yr of landfilling in Ämmässuo. In Kujala, the longer stabilisation time (around 40 yr) and greater amount of inert material, as well as landfill practices, had resulted in a low VS/TS ratio in the bottom layer. On the other hand, in Kujala, the present VS/TS (26%) was higher than the 20% VS/TS found at the lowest 12 m in the late 1980s (Ettala et al., 1988), which could suggest that leaching of organics from the upper layers may increase VS/TS ratio in the bottom layers. This was supported also by the fact that the leaching of COD showed only a slight depth-related decrease, while the VS/TS decreased more significantly (from top to bottom) in Kujala. In both landfills, the major source of organic (biodegradable) material (VS) is MSW. In studies evaluating MSW decomposition rates in landfills (Bookter and Ham, 1982; Jones et al., 1983; Suflita et al., 1992; Ham et al., 1993; Wang et al., 1994; Mehta et al., 2002), the cellulose content, cellulose-to-lignin or cellulose-to-VS ratios have been used as an indicator of the extent of degradation. Cellulose-to-lignin ratios of <0.2 in 30-yr-old waste (Bookter and Ham, 1982) and a ratio as low as <0.02 in waste landfilled 10–20 yr ago (Wang et al., 1994) have been reported. Bookter and Ham (1982) concluded that waste with a cellulose-to-lignin ratio of <0.2 and VS/TS ratio of <20% indicated relatively well-stabilised waste compared to less degraded waste with a cellulose-to-lignin ratio 0.9–1.2 and fresh waste with a ratio of 4. In the present study, a VS/TS ratio of 16%, indicating well-stabilised waste, was found only in the bottom layer in Kujala. The fact that not only biodegradable (e.g., cellulose), slowly biodegradable (e.g., lignocellulose) and recalcitrant material (e.g., plastics), but also inert material (e.g. ash, soil) contributes to the TS (and VS) of the sample and caused the high variation in VS/TS ratios, also at the lowest layers in both landfills.

In Ämmässuo, the 3-fold higher BMPs in middle layer than in two middle layers in Kujala were apparently due to differences in landfilled waste characteristics, as the age of the waste in these layers was similar in both landfills. The highest BMPs (160–180 m^3/t TS) in Ämmässuo were approximately half that of food waste (301 m^3/t TS, Eleazer et al., 1997), and higher than reported for grey waste (46–101 m^3/t TS; Jokela et al., 2001, 2002), suggesting high methane potential despite the fact that the waste was mainly from a period (1995 onwards) when source segregation was increasingly being implemented in this region. As expected, the BMPs were lowest in both landfills in the bottom layer, close to or below the water table, also suggesting that at that level waste had probably biodegraded, that biodegradable compounds had leached from the waste, or that little biodegradable material had originally been landfilled (Kujala). As a point of reference, the German landfill ordinance (AbfAblV, 2001) requires that waste in mechanical biological treatment should be stabilised to a level such that its gas production potential as measured by a standardised method (GB21) should be <20 Nl/kg TS over a period of 21 days. On the basis of the present BMP assays (the method has some differences compared to GB21 test) and the assumption that 50% of biogas production would have been carbon dioxide (not measured), only 13 samples (out of 32) from Ämmässuo and 5 samples (out of 11) from Kujala would have attained (data not shown) the maximum permissible value (<20 Nl/kg TS), indicating that waste had not yet been stabilised to this standard in either landfill.

The batch assays showed that water addition may speed up methane production, which is likely due to the dilution of inhibitory compounds or/and better transport of substrates to micro-organisms. However, the effect of water addition on methane production varied and was not depen-

dent on the sample's initial moisture, VS/TS ratio or BMP. Bogner (1990) screened 18 samples (VS 8–70%) from three US landfill sites and found that water addition (200% of dry weight) more than doubled the gas production from most samples. Gas production was higher for samples with low VS and high soil-refuse ratios. Thus it seems that the short-term effects of water addition are difficult to predict, while in general leachate recirculation is the main tool in the bioreactor operation of a landfill (e.g., Reinhart and Townsend, 1998; Kim and Pohland, 2003). However, as expected, the major stimulation to methane production in the batch assays was achieved with the addition of inoculum, which in practical conditions has been considered by adding inoculum to recirculated leachate (Bae et al., 1998). On the other hand, it must be noted that in the batch assays inoculum, besides providing micro-organisms, also provides nutrients and adjusts the moisture and their separate effects have seldom been studied. On the other hand, leachate recirculation, besides adjusting moisture, also transports substrates, nutrients and micro-organisms into the landfill body, all of which may favour methane production more than added moisture alone (Hartz and Ham, 1983; Barlaz et al., 1990).

In both landfills, the pH was within a range (6.7–8.9 and 7.0–8.2 at Ämmässuo and Kujala, respectively) enabling methanogenesis, although a smaller range (6.8–7.4) has been determined for optimum methanogenesis (Barlaz et al., 1990). The lower pH in the top layers of landfills (Ämmässuo, Jones et al., 1983) is apparently due to the accumulation of organic acids on account of the low populations of methanogenic bacteria, which cannot utilize the acids at the same rate as they are produced in fresh waste (Barlaz et al., 1990). The Kujala landfill may have been too dry (mean TS 68%, bottom layer 73%) for optimal methane production, as optimal TS is <50% (Gurijala et al., 1993) or 45%, the latter determined for 15-yr-old landfill body samples from Ämmässuo (Jokela et al., 2001). However, the optimum moisture content in landfills might be lower than laboratory values due to the higher density and lower available porous space of the waste material in field conditions compared to laboratory conditions. The TS content in the bottom layers in Kujala seemed to be stable as the mean TS content (70%) measured in the two bottom layers in the present study was about the same as measured at the lowest 12 m of the same landfill in the 1980s (71% TS, Ettala et al., 1988).

The approximately 4-fold higher COD leaching in Ämmässuo suggests lower stabilisation of organic matter than in Kujala (Table 4). Leachable COD decreased slightly from top to bottom in both landfills except at the top layer in Kujala, where the leaching was close to that of the bottom layer, probably due to the introduction of source segregation of biowaste. The COD leaching of fresh biowaste is several-fold (8.2 kg COD/t TS) higher than that of grey waste (approximately 0.7 kg COD/t TS) (Jokela et al., 2002), while in the present study mean leaching was 19 kg COD/t TS in Ämmässuo and 6 kg COD/t TS in Kujala, thus indicating that the leaching potential of COD had increased during landfilling. Both landfills contained leachable COD in all layers, even in the bottom layer in Kujala where low organic material content was assumed (VS/TS ratio, 16%). On the other hand, the shaking leaching test probably overestimates the amount of the leachable COD owing to use of the small particle size (shredded <50 mm) of the material in the test, which may increase solubilisation, and also owing to the biodegradability of cellulose in lignocellulosic materials, which are normally recalcitrant in landfill conditions (Stinson and Ham, 1995). On the other hand, in landfill conditions, part of the COD, which is considered as COD leaching potential in the present study, would probably be degraded biologically.

Nitrogen which, as ammonium, is often a leachate pollutant of long-lasting concern (e.g., Kylefors et al., 2003) was present in similar concentrations in both landfills (TKN content 2.4 g/kg dry weight in bottom and 3.5–5.2 g/kg dry weight in other layers). For comparison, the TKN content in waste at these two landfills was lower than that reported for separate waste fractions as 6.5–27 g/kg dry weight in a mechanically sorted organic fraction of MSW (Mata-Alvarez et al., 1993), 12 g/kg dry weight in grey waste (Jokela et al., 2002), 15–60 g/kg dry weight in sludges from wastewater treatment (Metcalf and Eddy Inc., 1991) and 4–12 g/kg dry weight in mechanically biologically treated MSW (Cappai et al., 2005; Boni et al., 2006). The similarity in the nitrogen content in the bottom layers suggests that enhanced ammonification below the water table reduced the amount of nitrogen during the landfill histories (approx. 20 and 40 yr). In the top layer, in Kujala the landfilled waste probably contained less nitrogen during the last few years owing to the source segregation of biowaste and the fact that sludges were no longer landfilled.

The NH_4–N leaching and NH_4–N leaching/TKN ratios in Ämmässuo were about twice those at Kujala despite the similar TKN content (g N/kg dry weight) at both landfills. This may indicate that nitrogen (of which proteins constitute the main source in waste) probably originated more from biowaste in Ämmässuo than in Kujala, as it has been shown previously that the NH_4–N leaching potential of fresh biowaste is higher than that of fresh grey waste (Jokela et al., 2002). The present NH_4–N leaching potential in the bottom and middle layers in Ämmässuo appeared to be high, despite the fact that the nitrogen content was highest in the upper layer. Ammonification and the leaching of nitrogen were apparently enhanced in the layers below or close to the water table; thus leachate recirculation to nitrogen-rich sectors or layers where leaching has been limited could probably reduce the time needed to ammonify and solubilise nitrogen. In estimating NH_4–N leaching, it must be noted that shaking leaching tests can be used to estimate the leaching potential of NH_4–N in the present situation, but for long-term estimations, the shaking leaching test is less applicable as it does not take into account biological

stabilisation, which in the long-term may have a strong influence on the leaching of NH_4–N (Kylefors et al., 2003). Moreover, the L/S ratio of 10 used in the shaking leaching tests may overestimate NH_4–N leaching because in field conditions a L/S ratio of 10 will be reached only after very long periods of time.

From a practical point of view, the empirical data obtained in the present study appears to be case- and target-dependent, and can be used only to assist planning for gas collection and leachate recirculation systems, as well as for estimating the nitrogen loads and energy potentials of the two landfills.

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II

INTERNAL LEACHATE QUALITY IN A MUNICIPAL SOLID WASTE LANDFILL: VERTICAL, HORIZONTAL AND TEMPORAL VARIATION AND IMPACTS OF LEACHATE RECIRCULATION

by

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Internal leachate quality in a municipal solid waste landfill: vertical, horizontal and temporal variation and impacts of leachate recirculation

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Abstract

The aim of this study was to monitor and characterise internal leachate quality at a Finnish municipal solid waste landfill (Lahti; Kujala, in operation for approximately 50 years) to provide information about its horizontal and vertical variation as well as effects of leachate recirculation on leachate quality. The study area (approximately 4 ha) of the landfill was supplied with 14 monitoring wells for leachate quality monitoring over 2-year period. The leachate was monitored for COD, BOD, TKN, NH_4-N, Cl, pH and electric conductivity. The results showed high horizontal and vertical variability in leachate quality between monitoring wells, indicating that age and properties of waste, local conditions (e.g., water table) and degradation and dilution processes have a marked effect on local leachate quality. The mean COD values (642-8037 mg/l) and mean BOD/COD ratios (0.08-0.17) from the different monitoring wells were typical of landfills in the methanogenic phase of degradation. The leachate in the monitoring wells was notably more concentrated than the leachate effluent used for leachate recirculation. In the landfill as a whole the effects of the leachate recirculation on leachate quality, although difficult to distinguish from those caused by other factors, appeared to be minor during the study period.

Key words: Landfill, leachate, sampling, organic matter, nitrogen.

1 Introduction

Landfilling has been the main method of municipal solid waste (MSW) disposal in most countries for last few decades. The composition of waste deposited in landfills is mainly determined by the consumption habits and waste management systems of the society in question as well as by changes in commonly used materials (e.g., introduction of plastics in the 1960s). In addition to MSW, municipal landfills often contain daily cover materials (e.g., surplus soils and composted sludges), which are used to minimise gas emissions (e.g., methane and odours) and leachate volume as well as littering in the local environment. Furthermore, in many landfills during their period of operation (typically 20-30 years) some areas have been used for the disposal of specific materials such as sewage sludge, ash, asbestos material, or dead animals. Thus, landfills are typically highly heterogeneous mass storage sites, where the deposited materials are commonly compacted to have a density varying from 0.8 to 1.0 t/m^3 [1].

In landfills a proportion of the biodegradable organic compounds are hydrolysed, acidified and subsequently methanised into the landfill gas (methane and carbon dioxide and trace components) within a few decades [2]. In landfills, biodegradation is often considered to be limited by low water content and slow leachate flow within the landfill, and thus leachate recirculation is a basic tool in the bioreactor concepts, which aim to control and enhance stabilisation of the landfill [e.g., 3, 4, 5]. Water, which enters the landfill within waste and as precipitate, and/or is actively introduced into the landfill, transports substrates and inhibitory compounds within the landfill body and leaches out organic compounds and non-organic compounds as well as chloride and metals. Excess water (landfill leachate) is collected, e.g., through a drainage system at the landfill bottom, and then treated either on-site or discharged into local sewage systems. Leachate quality is dependent on the stages of degradation of the waste in the landfill and contains varying amounts of organic matter (e.g., measured as chemical oxygen demand (COD), range 140 - 152 000 mg/l), inorganic macrocomponents (e.g., ammonium-nitrogen (NH_4-N) range 50 - 2 200 mg/l; chloride (Cl) range 150 - 4 500 mg) and smaller amounts of heavy metals as well as other organic or inorganic compounds [2].

In order to evaluate the post-closure needs (e.g., leachate quality) and / or to enhance the bioreactor operation of the landfill, information about the conditions inside the landfill body is required. Monitoring wells placed in the landfill body can be used to characterise the leachate quality at that point. This in turn could illustrate the conditions in the close vicinity of the well, such as the degradation stage (acidogenic or methanogenic) of the waste and its leaching potential as well as water movement in the landfill. Monitoring wells with screens enabling water to be gathered from different depths of the landfill can be used to obtain information about vertical conditions. However, it must be noted that inside the landfill body water may flow in preferential ways [6].

The flow channels and rates are affected by the landfill material and compaction. Hydraulic conductivity values from 10^{-2} (non-compacted) to 10^{-4} m/s (compacted) have been reported for MSW landfills [7]. The representativeness of leachate samples from a monitoring well, possibility for generalisation and factors affecting leachate quality are poorly known, and they are probably highly case-dependent.

Leachate quality at landfills has been widely studied [8, 9, 10, 11, 2], and several leachate recirculation or bioreactor landfill studies have additionally investigated the effects of leachate recircirculation on leachate quality [3, 5, 12]. However sampling methods varied (e.g., number of monitoring wells or sampling at leachate lagoons) and most studies have been based on only a few leachate samples from within a landfill or leachate from the landfill as a whole. On the basis of the previous studies (cited above) more detailed studies monitoring leachate characteristics and their short- and long-term variation as well as the effect of leachate recirculation on leachate quality are needed to estimate the representativeness of leachate samples. Due to the large size and heterogeneity of most landfills, and for economic reasons, internal leachate sampling must be coordinated with the available information on waste age and composition as well as local conditions at the landfill site.

The objective of this study was to evaluate the vertical and horizontal variation as well as the temporal variation in the internal leachate quality of a MSW landfill over a 2-year monitoring period. In addition to the effect of leachate recirculation on leachate quality was assessed. The study was performed in the case of a landfill which had been operated for about 50 years. For the purpose of the study 14 monitoring wells were installed in an area of approximately four hectares.

2 Materials and methods

2.1 Sampling sites and sampling

The study was performed in the MSW landfill of Kujala (Lahti, Finland), which contains 3.1 million tons of waste and has been operated for approximately 50 years, as previously described by Sormunen et al. (2008). The leachate from the whole landfill is collected by drains around the landfill, conducted to the leachate lagoon and either treated on-site or (excess leachate) pumped to a municipal waste water treatment plant for treatment. In the landfill a leachate recirculation area (approximately 1.2 ha) as well as a control area (approximately 3 ha) was established (Fig. 1.) The height of the landfill varied in the leachate recirculation area from 19.6 to 25.3 m and in the control area from 8.6 to 24.8 m. During the last 10 years from 10 to 15 m of MSW has been landfilled in the leachate recirculation area and in the control area on its north-west side, while the lowest 5 m in the whole study area dates back to the 1960s.

The leachate (temperature 10-22 °C) was recirculated in the leachate recirculation area from June to October in 2003-2005. The recirculated leachate was pumped from a leachate recirculation well (referred to as leachate well) to the leachate recirculation canals (codes 1 and 2, length 200 m) located in parallel (distance 50 m) across the leachate recirculation area. The leachate recirculation alternated between the canals in every 5-7 days. The recirculation rates were 12 m^3/d and 24 m^3/d and the total annual amounts during the three periods varied from 670 to 970 m^3 and from 1190 to 1450 m^3 for canals 1 and 2, respectively.

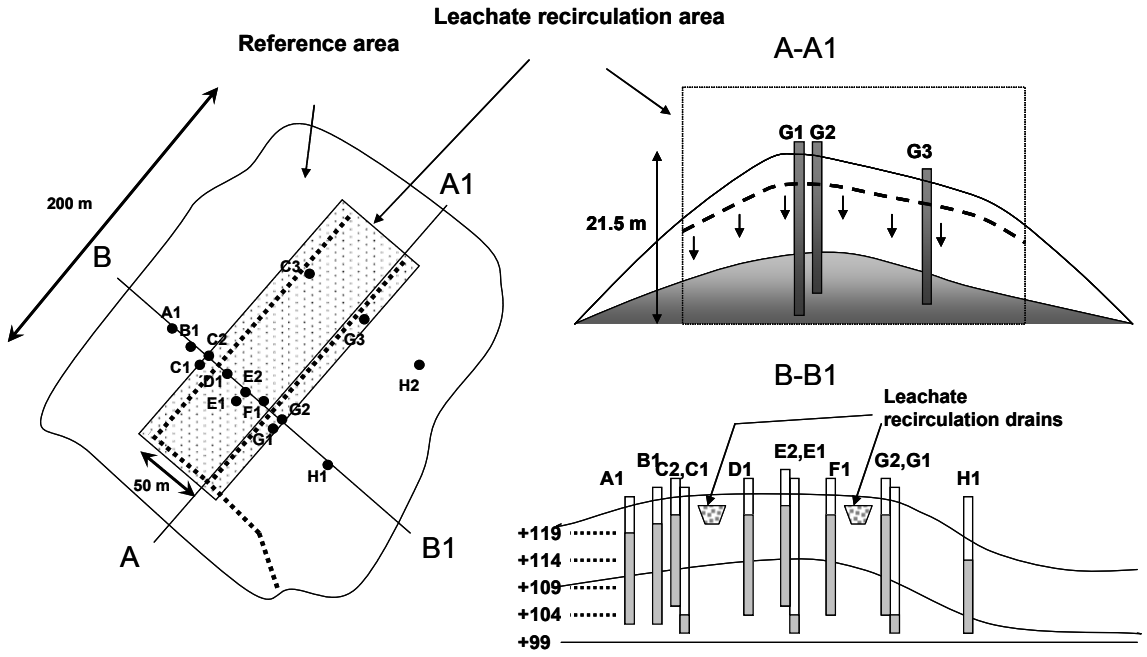

FIGURE 1 Sketch of the study area and locations and profiles (screens and extensions) of monitoring wells in the study area. Wells (e.g., A1, B1, C1) are shown in the picture (on the left) and cutaway pictures (A-A and B-B) on the right. The levels (Cutaway B-B) of the landfill bottom, levels of monitoring wells and their screens and extensions (right) were measured from mean sea level.

2.2 Monitoring wells

The fourteen monitoring wells (inner diameter 50 mm) were installed of which ten (C1, C2, D1, E1, E2, F1, G1, G2, C3 and G3) were located in the leachate recirculation area at a distance of approximately 5-25 m from the leachate recirculation canals, while the four monitoring wells (A1, B1, H1, H2) were located in the control area at approximately 25-60 m from the leachate recirculation canals. The monitoring wells were horizontally located at intervals of 5 to 100 meters and vertically located so that the bottom of the monitoring wells were situated at varying levels up to 6 meters from the bottom. The 11

monitoring wells were equipped with a long screen (varying from 6.5 to 21 m) with a short (3-4 m) extension leading to the top of the landfill. Wells C2, E2 and G2 (with varying screen lengths from 16 to 18 meters) were installed in parallel (distance 5 meters) with wells C1, E1, G1 (screen length 2 meters) to obtain samples from the upper leachate at 3.5 to 6 m distance and close (< 1 m) to the bottom of the landfill. The leachate was sampled in the monitoring wells and in the leachate (recirculation) well by bailers (Clear-View, high density polyethylene, volume 1 l). Approximately 2-3 liters of leachate were drawn from the monitoring wells before the samples for the analyses were taken.

2.3 Analysis and measurements

COD, biological oxygen demand (BOD) and total Kjeldahl nitrogen (TKN) as well as NH_4-N were analysed according to SFS standards [13, 14, 15]. The samples for the NH_4-N and COD were filtered with glass fibre filter papers (GF50, Schleicher and & Schuell), except for the samples taken in 2005, which were not filtered. A Hanna Instruments (Hi9025) pH meter and a Sensorex pH 450 CD electrode were used to measure the pH of leachate immediately after sampling in 2004. A Hanna instruments (Hi 9635) meter was used for the conductivity measurements. The water tables were measured by a sensor (Labko 4390-027) in some of the monitoring wells (C1, C3, G2, G3, H1; Fig. 1) with on-line data collection.

3 Results

3.1 Leachate characteristics and vertical profile

Table 1, which summarises (mean ± std) the leachate characteristics in the different monitoring wells (Fig. 1) and in the leachate recirculation well during the 2-year study period, shows that the mean concentrations and values of the studied parameters varied between different wells and also that variation occurred within most of the wells during the study (Fig. 2 and 3). For example, the lowest and highest individual COD values measured were about 400 mg/l and 10 000 mg/l, while the lowest and highest mean values in different wells were 642 ± 46 mg/l (E1) and 8037 ± 1138 mg/l (E2). In general, the differences between different wells were much higher (as percentages) in COD and BOD than in the other parameters (e.g., NH_4-N, Cl).

The leachate characteristics were studied as a function of the level of the monitoring well (gathering water from low or high levels of the landfill; Fig. 1, 4).

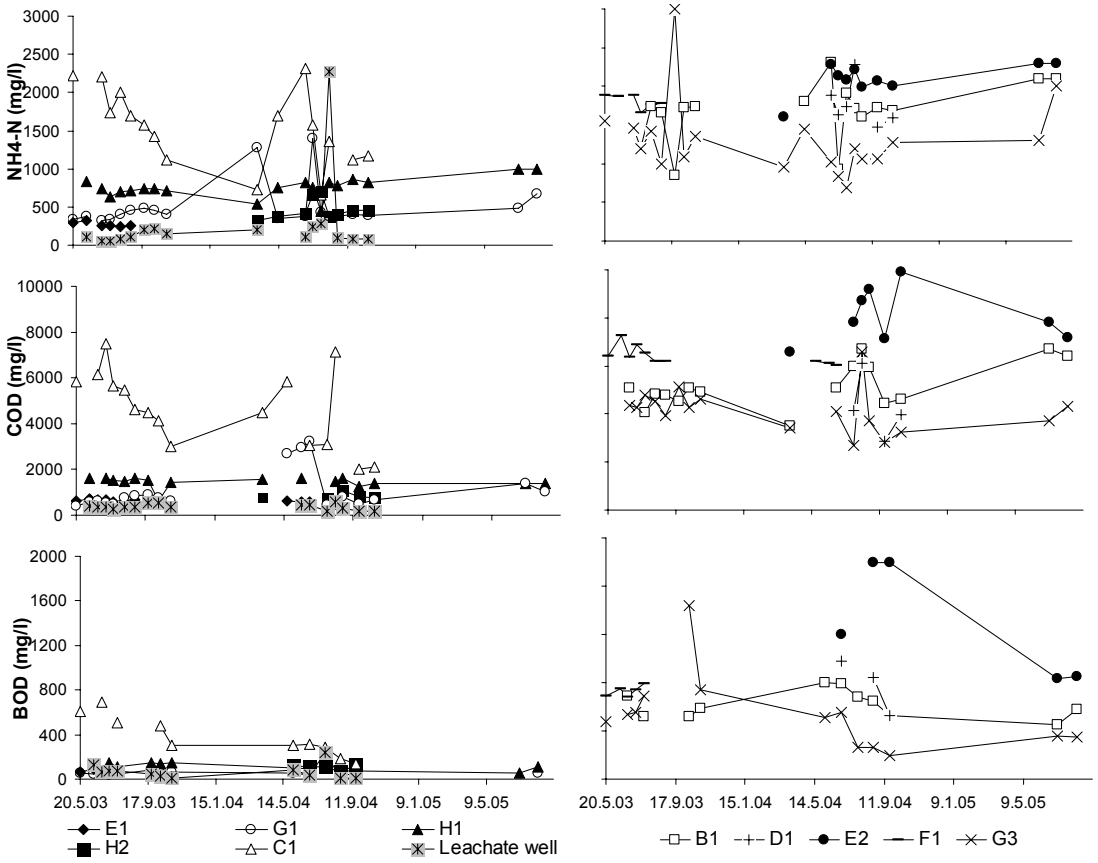

FIGURE 2 NH_4-N, COD and BOD in low (left) and high (right) level monitoring wells and in leachate (recirculation) well.

The mean concentrations or values of most parameters in the leachate recirculation area were lower in the two low level monitoring wells (E1, G1) than at higher levels, although one low level well (C1) also had higher values. On the other hand, low values were observed also in high level wells in the reference area (H1, H2). The highest NH_4-N concentrations and COD as well as BOD values were observed in well E2, which, however, had a well profile comparable to that of most of the wells (e.g., B1, D1, F1 and G3). Moreover E2 also had significantly higher values compared to E1, located only 5 m away from it but gathering leachate from the lower level of the landfill (E1 99.5-101.5 m, E2 104.94-122.94 m). The BOD/COD (means from 0.08 to 0.17 in different monitoring wells) and NH_4-N/N (means from 0.75 to 0.90 in different monitoring wells) ratios as well as pH (from 6.9 to 8.5) did not vary significantly with landfill level.

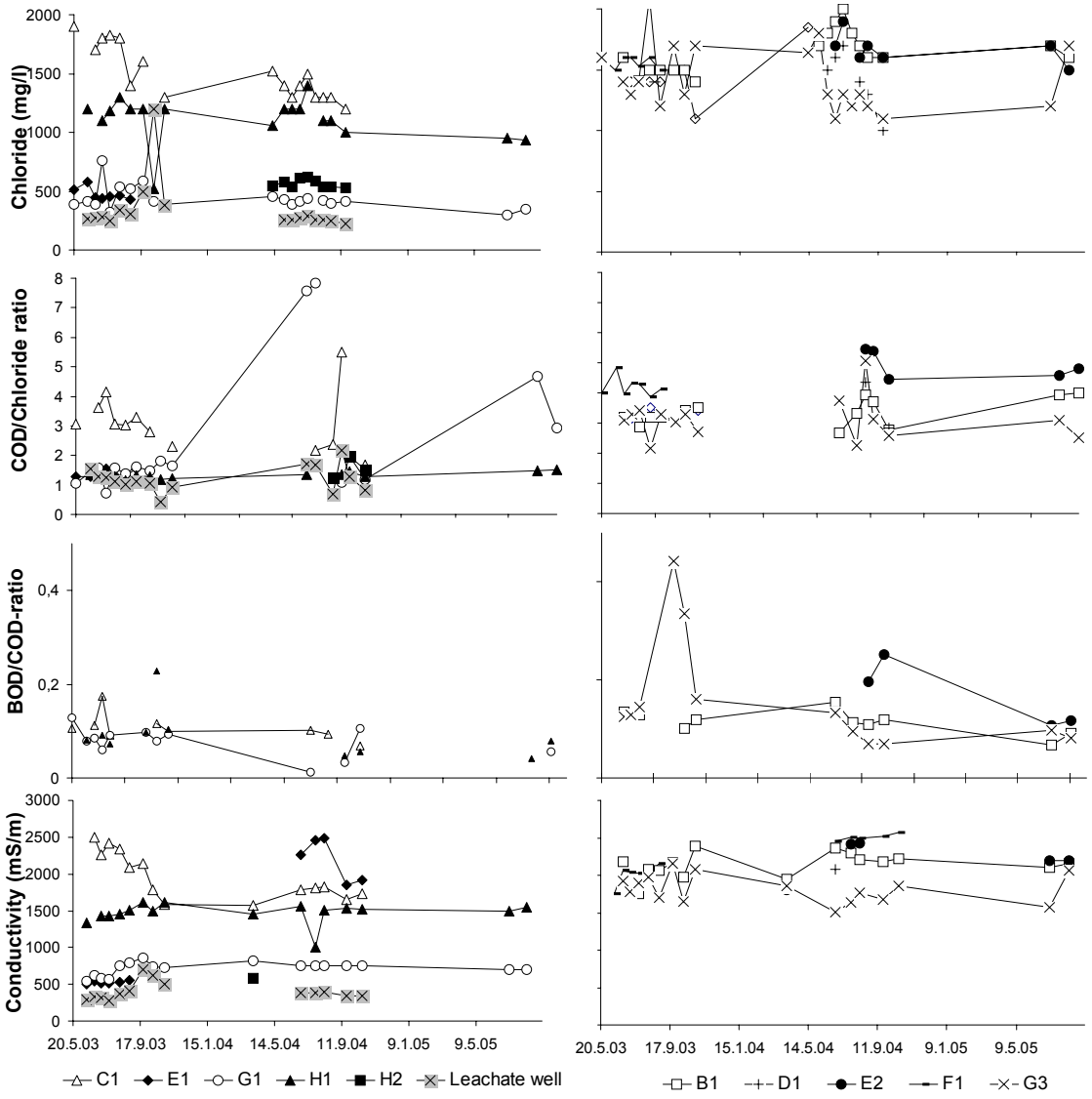

FIGURE 3 Chloride, COD/Chloride and BOD/COD ratios and electric conductivity in low (left) and high (right) level monitoring wells and in leachate (recirculation) well.

3.2 Effect of water table and sampling depth on leachate quality

The water table (measured from mean sea level) varied between the monitoring wells (from +102.6 to 103.7 in H1; from +103.8 to +103.9 in C1; from +108.7 to 109.0 in C3; from +110 to +110.7 in G2 and from +104.0 to 104.9 in G3). This probably had an effect on leachate quality via sampling depth as the mean zone of saturation varied from approximately 20 % (in C1) to 51 % in (G2) of the height of the landfill, which meant that the screens of the low level monitoring wells (C1, E1, G1; with short screen 2 m) were in the saturated part while the

screens of the higher level monitoring wells (with long screen from 6.5 to 21 m) were partly in the non-saturated part.

3.3 Temporal and horizontal variation in leachate quality

The concentrations in the monitoring wells at varying distances (from 5 to 25 meters) from the leachate recirculation canals varied randomly and were not dependent on the distance of the monitoring well to the recirculation canal. However, the variation in the COD/chloride ratio in the two low level monitoring wells (C1 and G1) and in the higher level well (G3), located at 5 meters distance from the leachate recirculation canal might indicate that the recirculated leachate was diluting the chloride content in the leachate in the landfill body (Fig. 3), as the concentrations of chloride in the recirculated leachate, which was collected from the whole landfill area, were lower (252-500 mg/l, mean 345 mg/l) than in the leachate in the landfill body (300-2100 mg/l, mean 1217 mg/l). However, the leachate quality in G1 varied less than in monitoring wells C1 and G3 on the basis of the electric conductivity, which may also indicate local effects of leachate recirculation on leachate quality in certain monitoring wells.

TABLE 1 Landfill height at locations of monitoring wells, screen range of monitoring wells and characterisation (mean±stdev, except pH range) of leachate samples.

Well	Height[a] (m)	Screen range (m) above mean sea level	Distance to recirculation canals	Conductivity (mS/m)	pH[b] (range)	NH_4-N (mg/l)	TKN (mg/l)	COD (mg/l)	BOD (mg/l)	Cl (mg/l)	BOD/COD ratio
C1	24.6	99.9-101.9	5	1963±317	7.4-8.1	1536±501	1863±497	4651±1681	338±182	1503±226	0.11±0.03
C2	25.1	104.1-122.1	5	na	7.3-7.7	1352±275	1968±668	na	na	1357±75	na
D1	25.3	103.3-122.3	5	2080	7.8-8.1	1765±288	2260±498	4269±1348	774±230	1417±248	na
F1	22.5	103.1-119.1	5	2235±279	na	1809±91	2207±133	6422±389	748±45	1561±49	0.11±0.007
G1	21.5	98.7-100.7	5	716±88	6.9-7.2	505±294	479±79	1030±862	53±14	436±102	0.08±0.03
G3	19.6	101.5-115.2	5	1819±193	7.3-7.8	1311±503	1457±448	4152±909	580±319	1427±271	0.16±0.11
E1	24.4	99.5-101.5	25	1282±891	7.9-8.1	271±31	286±85	642±46	75±10	475±53	0.11±0.01
E2	24.6	104.9-122.9	25	2206±256	7.7-8.5	2099±209	2553±308	8037±1138	1301±483	1671±125	0.17±0.07
B1	24.8	102.0-120.0	25	2139±165	7.6-7.9	1676±391	2082±520	5172±931	614±114	1636±169	0.11±0.02
A1	22.4	102.4-119.4	45	2113±102	na	1835±211	2329±536	4427±489	494±106	1442±242	0.11±0.04
H1	13.2	99.9-109.9	55	1468±143	7.4-7.7	757±132	881±134	1446±245	106±33	1113±188	0.09±0.05
H2	8.6	100.8-107.3	60	na	na	464-130	427±82	822±136	105±24	567±34	0.12±0.05
LW[c]				402±122	7.2-8.7	273±540	131±54	363±121	1310±66	344±230	0.13±0.11

[a]Height of the landfill body, [b]2004 data, [c]LW (Leachate well)

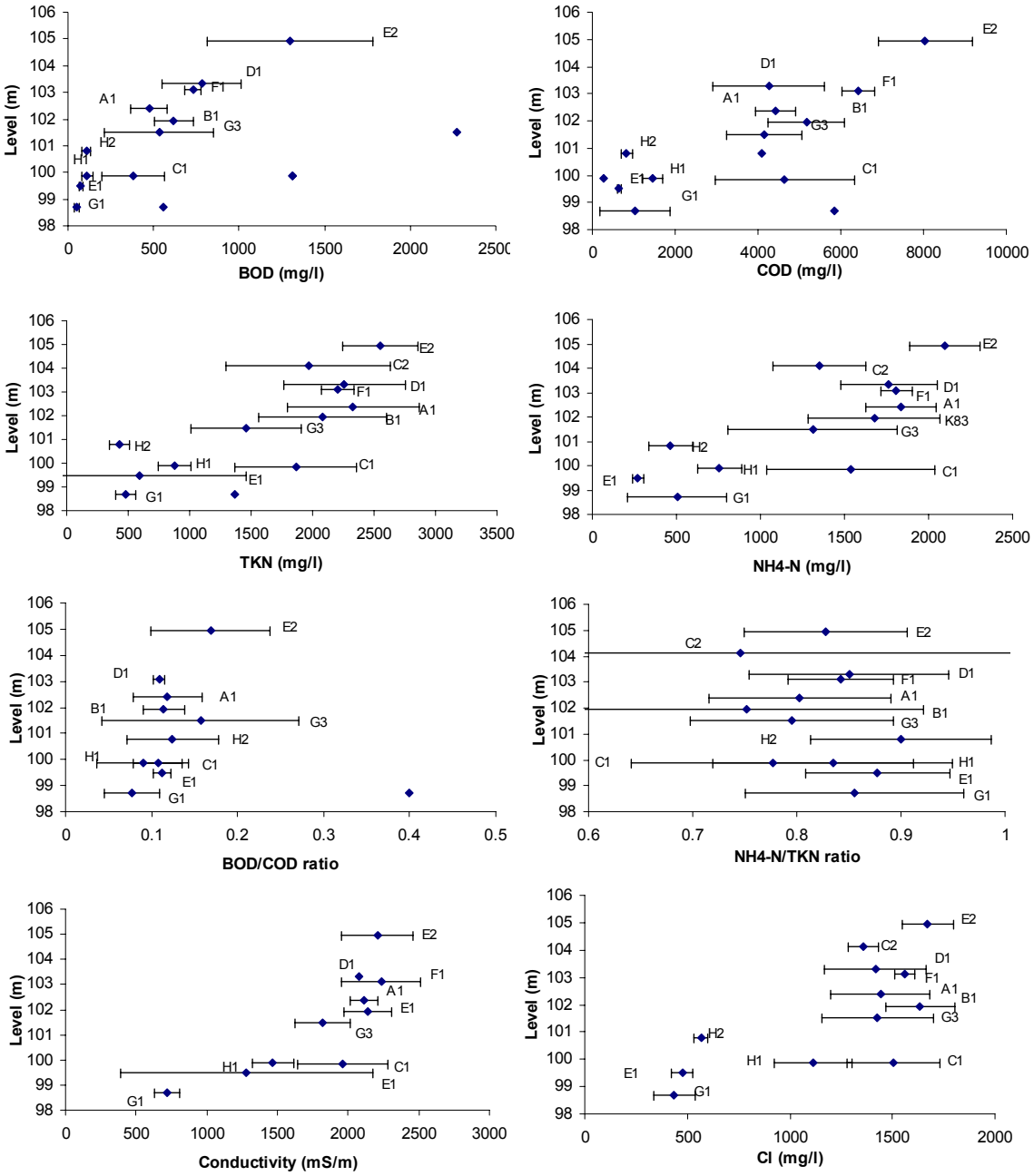

FIGURE 4 Mean concentrations of BOD, COD, N and NH_4-N, BOD/COD and NH_4-N/N- ratios, as well as mean conductivity and chloride concentrations in leachate obtained from monitoring wells located at different levels in the landfill body.

4 Discussion

4.1 Internal leachate sampling

The present study shows the possibilities of internal leachate sampling to characterise leachate quality within a MSW landfill that has been in operation for several decades. In particular, it seems that leachate quality can vary considerably both vertically and horizontally, as found in the present samples from monitoring wells located at a distance of only 5 to 150 m from each others. It is therefore evident [11, this study] that reliable characterisation of leachate quality within a MSW landfill requires the systematic implementation of monitoring wells for sampling in saturated waste layers and water table measurements. Moreover waste quality at different areas and layers should be considered in planning the placement of wells as well as the lengths and screen levels of wells to take into account the effect of attenuation (e.g., dilution or degradation) processes on leachate quality. The minimum number of monitoring wells would seem to depend on the landfill properties (waste age and landfill filling procedure) as well as on the aim of the sampling, and thus no general recommendations can be given.

4.2 Horisontal and vertical variation in leachate quality

The variation in leachate quality between monitoring wells with a similar vertical profile and located at the same level from the bottom (e.g., G3 and H2, which had 3-fold differences in NH_4-N and 5-fold differences in COD) could be due to initial spatial differences in waste characteristics and / or differences in stages of degradation (e.g., acidogenic or methanogenic) as well as differences in the level of the water table. The variation in leachate quality may also be due to differences in the local hydraulic conductivity of the waste and of the daily cover layers, which may greatly impact on water flows inside the landfill body causing preferential pathways for leachate [6, 16]. The present study shows that in general the leachate was more concentrated in the study area, where the waste was fresher (<10 years in the upper parts), than in the south-east parts of the control area, where landfill height was lower and the waste older (>20 years). This indicates that the waste was more stabilized in the south-east parts of the control area either through methanisation and /or due to leaching with time, as one would expect.

In the studied landfill the leachate at the bottom (up to 2 - 3 meters from the bottom) was strongly (3-8 fold) diluted compared to that at higher levels (from 4 to 7 meters from the bottom) as indicated, e.g., by the fact that the COD value and chloride concentration decreased towards the bottom. The decrease in COD values alone could indicate methanisation of COD with landfill body depth, while chloride is assumed to be useable as a tracer compound due to its inert nature [16, 17]. In the present landfill, the dilution stream in the bottom

layers apparently proceeds either horizontally from more stabilized areas or as preferential pathways through the landfill, or it may be due to groundwater in the landfill in the absence of bottom structures. Unfortunately it was not possible to verify either of these dilution routes on the basis of existing information on the landfill. However, the present study suggests that leachate quality may show significant variation vertically, especially, it would seem, if the water saturated part of the landfill is high, as in the present case, where from 20 to 51 % of the landfill height was saturated.

4.3 Effects of leachate recirculation on internal leachate quality

It was not possible to differentiate and observe the short-term effects of leachate recirculation on internal leachate quality in the present 50-year-old MSW landfill, despite the fact that, the leachate quality varied most in the monitoring wells in close proximity to leachate recirculation canals. This may be because the variation in leachate quality due to leachate recirculation is low and /or because the variation in leachate quality is affected by other factors such as degradation processes and water flows due to precipitation and/or melting snow. The latter was proven by the temporal variation observed in leachate quality in wells located in the lower parts of the control area, which probably had the most stabilized waste. Moreover the horizontal distribution of the recirculated leachate may be limited to a few meters only, as described previously [18]. In the present landfill in particular, the horizontal distribution may be low, as low hydraulic conductivities from $6 * 10^{-5}$ to $5 * 10^{-4}$ m/s [19] were measured in the 1980s. In addition to low hydraulic conductivity the preferential leachate flows may reduce the effective area of leachate recirculation [16, 18]. Change in the stages of degradation may, in the present case, be indicated by the lower BOD/COD ratio in the leachate recirculation area than in the control area. Previous studies have shown that leachate recirculation does not have a marked effect on the BOD/COD ratio [5] and that leachate recirculation may even raise the BOD/COD ratio [12] during the first 2-3 years of leachate recirculation in a bioreactor landfill due to enhanced degradation in the acid phase.

4.4 Monitoring of landfills by leachate characterisation

The leachate in the landfill as a whole is apparently diluted by surface runoff or direct rainfall into the leachate lagoon, as indicated in this study by the fact that the leachate in the leachate well was more diluted than the leachate samples obtained from the landfill monitoring wells. Moreover the leachate may have been diluted by leachate collected from areas outside the present study area. The mean COD value (5610 mg/l) and the mean NH_4-N concentration (1732 mg/l) obtained from the higher level monitoring wells were higher than the reported means for landfills from 1 to 5 years old (3810 mg/l for COD and 405 mg/l for NH_4-N, respectively) [20]. Thus it is clear that the present landfill still

contains high amounts of easily transportable organic material and nitrogen, which flow through the landfill. The BOD/COD ratio (mainly 0.05-0.2) in most of the observation wells was typical of a methanogenic (0.02-0.24) landfill [2], indicating the degradation of organic matter within the landfill. Moreover pHs enabling methanogenesis were found in all the monitoring wells, even though fresh waste may cause an accumulation of organic acids, as previously described [21]. In the present case the population of methanogenic bacteria in the lower (15 m from bottom, landfilled in 1960-1997) part [22] of the landfill was probably high enough to utilize the organic acids produced in waste deposited in the higher parts (from 5 to 10 m from the top) within 5 years. Thus in this particular landfill it might be worthwhile to evaluate the possibility of recirculating the more concentrated leachate from the higher parts or elsewhere of the landfill instead of diluting the leachate of the whole landfill, as one of the aims of bioreactor operation is to recirculate the biologically degradable organic material back to the landfill, thereby promoting its degradation [23].

5 Conclusions

- In a landfill with a long history of operation internal leachate quality may vary greatly both horizontally and vertically. Moreover there may be temporal variation in leachate quality in both the lower and upper parts of the saturated landfill body.
- The leachate was most concentrated in the upper part of the leachate-saturated landfill body in locations where the water table was high and newly landfilled waste formed the top layers.
- The most diluted leachate was in the bottom part of the landfill; thus degradation and dilution processes seem to have a notable effect on the vertical profile of the leachate.
- The effect of leachate recirculation on internal leachate quality may be hard to isolate due to variation caused by other factors during short-term leachate recirculation.
- General recommendations about the number of observation wells in respect of the size of the study area cannot be given due to fact that local conditions, waste management history and landfill practices may have a considerable effect on the representativeness of leachate samples.

Acknowledgements

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III

LONG-TERM ON-LINE MONITORING OF WATER TABLE, TEMPERATURE AND LEACHATE QUALITY IN TWO MUNICIPAL SOLID WASTE LANDFILLS

by

Kai Sormunen, Matti Ettala & Jukka Rintala

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Long-term on-line monitoring of water table, temperature and leachate quality in two municipal solid waste landfills

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Abstract

The aim of this study was to evaluate the feasibility of internal on-line monitoring of water table, temperature, electrical conductivity and pH in municipal solid waste (MSW) landfills. The study was performed in test areas located in two Finnish MSW landfills (Ämmässuo and Kujala, in operation for approximately 20 and 50 years, respectively). Instrumented monitoring wells (four in Ämmässuo and five in Kujala) were established in the proximity of areas where leachate recirculation was practised during the summer. The results showed variation in water table (from 3.5 to 9.5 m and from 2.5 to 10.5 m measured from bottom stucture) and temperature (from 10 to 39 °C and 17 to 35 °C in Ämmässuo and Kujala, respectively) between the monitoring wells. Variations, mainly minor, were also observed in water table and temperature within the same monitoring wells during the approximately one and a half year study. The greatest variation in electrical conductivity (range 1300-2300 mS/m) occurred in a monitoring well located next to the leachate recirculation canal in Kujala. In general, variation in water table, temperature, electrical conductivity and pH was slow and clear seasonal variations were not observed, despite boreal conditions and the varying height of test areas (range 10 to 24 m and 12 to 25 m in Ämmässuo and Kujala, respectively). These limitations aside, the present study showed that on-line monitoring of internal leachate in MSW landfills is feasible and that automated data collection can be performed by wireless data transfer.

Key words: landfill, monitoring, on-line, leachate, water table, temperature.

1 Introduction

Municipal solid waste landfills are heterogeneous deposits of materials, in which conditions vary greatly. This has prompted discussion regarding the rate of biological stabilization as indicated by biogas production from organic materials. Recently, biogas has been recovered in increasing quantities due to fact that methane, a greenhouse gas, accounts for a major proportion of landfill biogas. Thus landfills are notable sources of greenhouse gas (IPCC 2006). On the other hand methane can be recovered and used as a renewable energy source to minimize pollution and the use of fossil fuels. Landfill bioreactors have been introduced in order to both promote degradation and to increase gas production in municipal solid waste (MSW) landfills (e.g., Benson et al. 2007, Yazdani et al. 2006). Traditionally, the main operation in landfill bioreactors has been leachate recirculation; however, within the last decade aeration of landfills has also been developed (Heyer et al. 2005). Depending on the waste quality, stage of degradation and conditions, leachate in MSW landfills is highly polluted, e.g., in terms of organic matter and nitrogen (Kjeldsen et al. 2002). Internal leachate within the landfill seems to have an important role in the processes of degradation and leaching (reviewed by Komilis et al. 1999; Sormunen et al. 2008) while excess leachate must be treated prior to release into the environment.

Due to the heterogeneous nature of a MSW landfill, spatial and vertical as well as short-term variation may exist in leachate quality, as found in a previous study (Sormunen et al. 2007). In young landfills leachate generation is typically low due to fact that infiltrated water may continue to accumulate still in 10 to 20-year-old landfills (Bengtson et al. 1994). With time, as the moisture level approaches to water-holding capacity, high rates of precipitation and melt waters may cause an increase in leachate generation, while higher evaporation from May to September minimizes leachate generation, at least in landfills located in temperate climatic conditions (Bendz et al. 1997). Thus seasonal fluctuations may exist in leachate generation and in the level of the water table in a landfill. Moreover, in addition to high water table a high pore water pressure may cause mechanical stability problems in a landfill with low water permeability as reported for landfills containing mechanically-biologically treated waste (Stegmann et al. 2005). Also landfill operation as a bioreactor may cause a rise in the water table and changes in leachate quality (Benson et al. 2007).

Changes in the water table and leachate quality have traditionally been monitored by sampling and laboratory analysis or field measurements, while internal on-line monitoring has rarely been studied, even if on-line monitoring (e.g., pH, electrical conductivity, COD, turbidity) has been widely used in wastewater quality monitoring (Thomas & Pouet 2005). However leachate quality may vary considerable even in same monitoring well (Sormunen 2007),

thereby causing uncertainty in the results obtained from traditional leachate quality monitoring methods.

The objective of this study was to study the feasibility of the on-line monitoring of the water table, temperature, electrical conductivity and pH in internal leachate quality monitoring. The main objectives were to assess the technical feasibility of these measurements and to assess the utility the data obtained.

2 Materials and methods

2.1 Study areas

The study was performed in the MSW landfills of Ämmässuo (Espoo, Finland) and Kujala (Lahti, Finland), which have been in operation for approximately 20 and 50 years, respectively. Simultaneously with the present study, waste characteristics (Sormunen et al. 2008) were studied in both landfills and leachate (Sormunen et al. 2007) characteristics in the Kujala landfill. In order to study the feasibility of on-line monitoring, four instrumented monitoring wells (inner diameter of 50 mm) were bored in Ämmässuo and five in Kujala. Moreover most of the on-line measurements were established in proximity to leachate recirculation canals to study effect of leachate recirculation on water table, temperature and electrical conductivity.

2.2 On-line measurements

2.2.1 Ämmässuo

Four instrumented monitoring wells (named A, B, C, D; depths between 8.5 and 21.5 m) for temperature and water table monitoring were bored in 2003 in one of the three landfilling areas (Sector 1) in Ämmässuo landfill (Figure 1, Table 1). Instrumented monitoring wells were bored on the landfill slope, where height of the landfill varied from 10 to 24 meters. Leachate recirculation (approximately 310 mm in 2003 and 460 mm in 2004) was practised from June to October in an area of approximately 0.5 ha. Monitoring wells A, B and D were in close proximity to the leachate recirculation canals, the distance from a monitoring well to the closest leachate recirculation canal varying from 5 to 18 m, while monitoring well C was located at approximately 80 m distance from the leachate recirculation area.

The monitoring wells were instrumented by a piezoresistive pressure transmitter (Keller PR-36W) for the water table and a thermistor (Betatherm 10K3A1B) for temperature. All the monitoring wells extended from 2 to 4 m from the bottom liner of the landfill. The monitoring wells were equipped with screens varying from 2 to 13 m and extensions from 4 to 22 m. Data were recorded hourly (for periods between 194 and 609 days in 2003-2004) in a

datalogger (Cambell CR10X) and transferred by a GSM modem (Siemens TC35i) with datalogger support software (Campbell Scientific, version PC208W) to a computer as required. The datalogger, GSM modem, accessories and battery were located in a mast-mounted measuring station on the landfill. The datalogger and GSM modem were powered by a solar photovoltaics panel installed outside the measuring station.

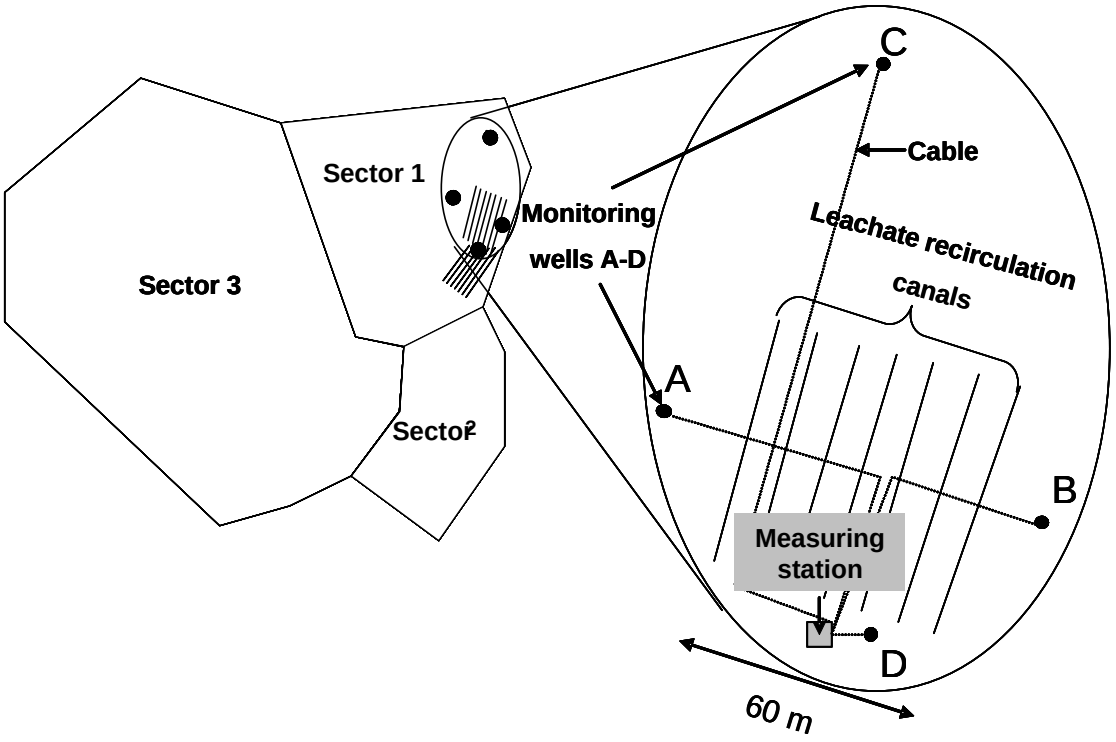

FIGURE 1 Location of on-line measurements of water table and temperature in monitoring wells (A, B, C, D) and leachate (seven horizontal canals) recirculation canals in Ämmässuo landfill.

2.2.2 Kujala

Five instrumented monitoring wells (named C1, H1, G2, G3 and C3; depths between 13 and 25 m) were bored in 2003 in Kujala landfill (Figure 2), where height of the landfill body varied from 12 to 25 meters. Leachate recirculation was practiced in close proximity to monitoring wells (5 to 60 m distances) from June to October (approximately 600 mm in 2003 and 500 mm in 2004) in an area of approximately 1 ha.

TABLE 1 Height of landfills in on-line instrumentation locations, and bottom as well as top levels of monitoring wells with their screens in Ämmässuo and Kujala landfills.

Ämmässuo	Height of landfill (m)	Length of monitoring well (m)	Bottom level (m)[a]	Top level (m)	Lengths of screen (m)
A	24	21.5	64.4	85.9	2
B	10	8.5	62.9	71.4	6
C	20	16	63.6	79.7	2
D	17	16	61.9	77.9	13
Kujala					
C1	25	25	99.9	124.9	2
H1	12	13	99.9	113.2	10
C3	24	24	100.5	124.5	21
G2	21	19	102.5	121.5	16
G3	18	17	101.5	118.2	14

[a]Levels of monitoring wells were measured from mean sea level.

For the on-line measurement of the water table and temperature in the monitoring wells a combined sensor (Labko 4390-027) for temperature and water table was installed in four monitoring wells (H1, G2, G3 and C3). Moreover on-line conductivity was studied in three monitoring wells (C1, G3 and H1; electrode GLI 3725E) and pH in one monitoring well (H1; electrode GLI PD1P1). The monitoring wells were equipped with screens between 2 and 21 m and extensions from 2 to 23 m (Table 1). The water table and temperature were monitored for varying periods between 212 and 516 days and conductivity (C1, G3 and H1) and pH (H1) for 167 days. Data transfer from electrodes (through logic control and a radiomodem) to the control room was continuous, and real-time data was displayed both on a field monitor located in the proximity to monitoring wells and in a monitor located in the control room. On-line data were saved once per hour in the control room computer. The measuring systems on the landfill were connected to an electrical network.

2.2.3 Comparison by field meters

The performance of the on-line conductivity and pH measurements was compared with that of manually conducted field measurements in Kujala for approximately five months. The monitoring wells were manually sampled, immediately after which conductivity and pH were determined on-site by the field meters, as previously described (Sormunen et al. 2007). The comparisons were done weekly for approximately one month, then twice monthly for two months and, finally once a month. The on-line conductivity and pH electrodes were calibrated from three to five times during the comparison period from 1st June till 12th October in 2004. Moreover from 31st August till 1st September the on-line monitoring of pH was tested to monitor a sudden fall in pH and the electrode was kept in buffer solution (pH 4) for a day.

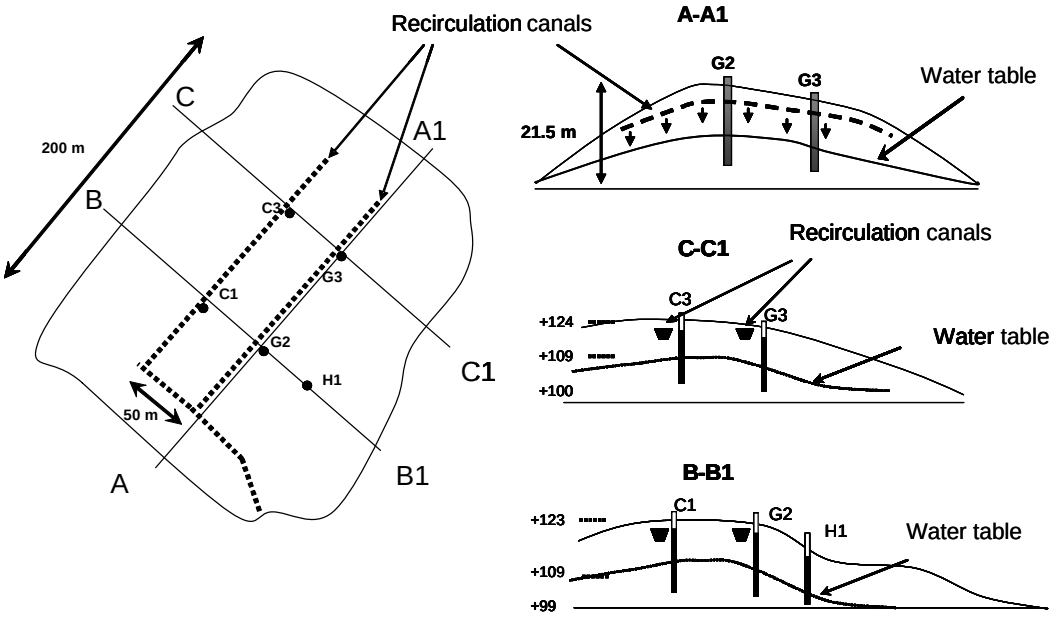

FIGURE 2 Location of on-line measurements: water table and temperature in H1, G2, G3 and C3; conductivity in H1, G2 and C1; pH in H1 and leachate recirculation (2 horizontal canals) in Kujala landfill.

3 Results

3.1 Comparison of on-line and manual measurements

The feasibility of monitoring electrical conductivity and pH of leachate by on-line measurements was studied in instrumented monitoring wells in Kujala landfill. The on-line pH values (H1) were consistently 0.2 to 0.3 higher during the 5-month period than the manual values, while both measurements showed similar trends (Fig. 3). On-line pH showed values between 4.3 and 4.4 due to fact that the electrode was in buffer solution (pH 4) outside of the monitoring well owing to testing for a sudden change in pH during 31.8-1.9.2004 and for calibration.

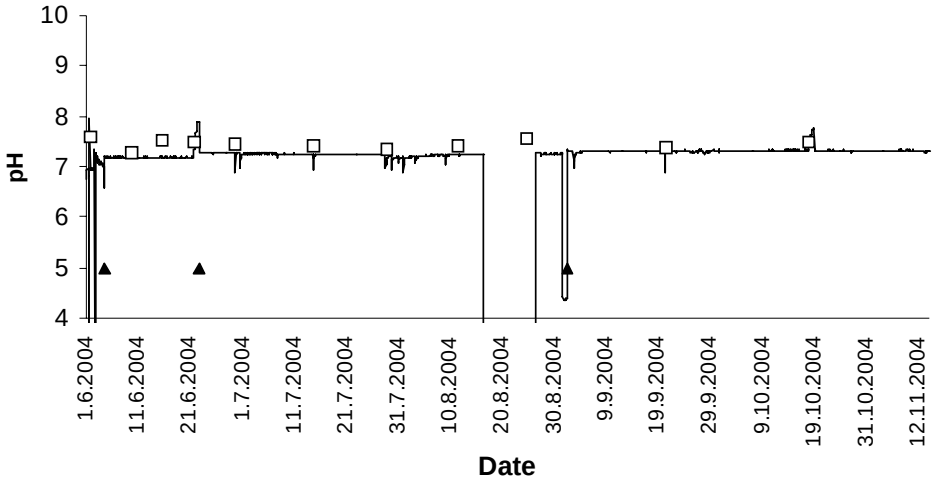

FIGURE 3 On-line (--) pH, calibrations (▲) and pH of leachate samples measured by field meter (□) at observation tube H1.

On-line and manual measurements of electrical conductivity were compared over a period of 5 months in three monitoring wells (Fig. 4; C1, H1, G3). During the study the on-line measurements differed by less than 100 mS/m in C1 (on-line from 1340 to 1970 mS/m; manual from 1660 to 1840, excluding the first three weeks) and, mostly, by less than 200 mS/m in G3 (on line from 1220 to 2330; manual 1220 to 2860 mS/m) and less than 50 mS/m in H1 (on-line from 1260 to 1550, manual from 1000 to 1560, excluding the first three weeks) until the third calibration (1.9.2004) caused a systematic decrease of approximately 200 mS/m in the on-line level of conductivity compared to the values obtained by the field meter.

3.2 Water table and temperature monitoring in Ämmässuo

Variation and seasonal changes in the water table and temperature were studied by on-line measurements in four monitoring wells (A, B, C, D) in Ämmässuo landfill for periods between 194 and 609 days (Fig. 5). The highest and lowest measured water table and temperature in the four monitoring wells ranged from 3.5 to 9.5 m and from 10 to 39 °C, respectively. The water table varied within two meters in same monitoring wells during the study. The water table increased gradually in B, C and D, while in A the water table fell by 1.5 m during two months (April and May) in 2003 and varied thereafter within 0.5 m during the next ca 14 months. The highest variation in temperature of 12 ° C (from 39 to 27 ° C) was observed with a decreasing trend in A, while in B the temperature varied within 8 ° C (from 12 to 20 ° C) without showing a clear trend and in C and D temperature varied within 2 ° C, in the latter with an increasing trend.

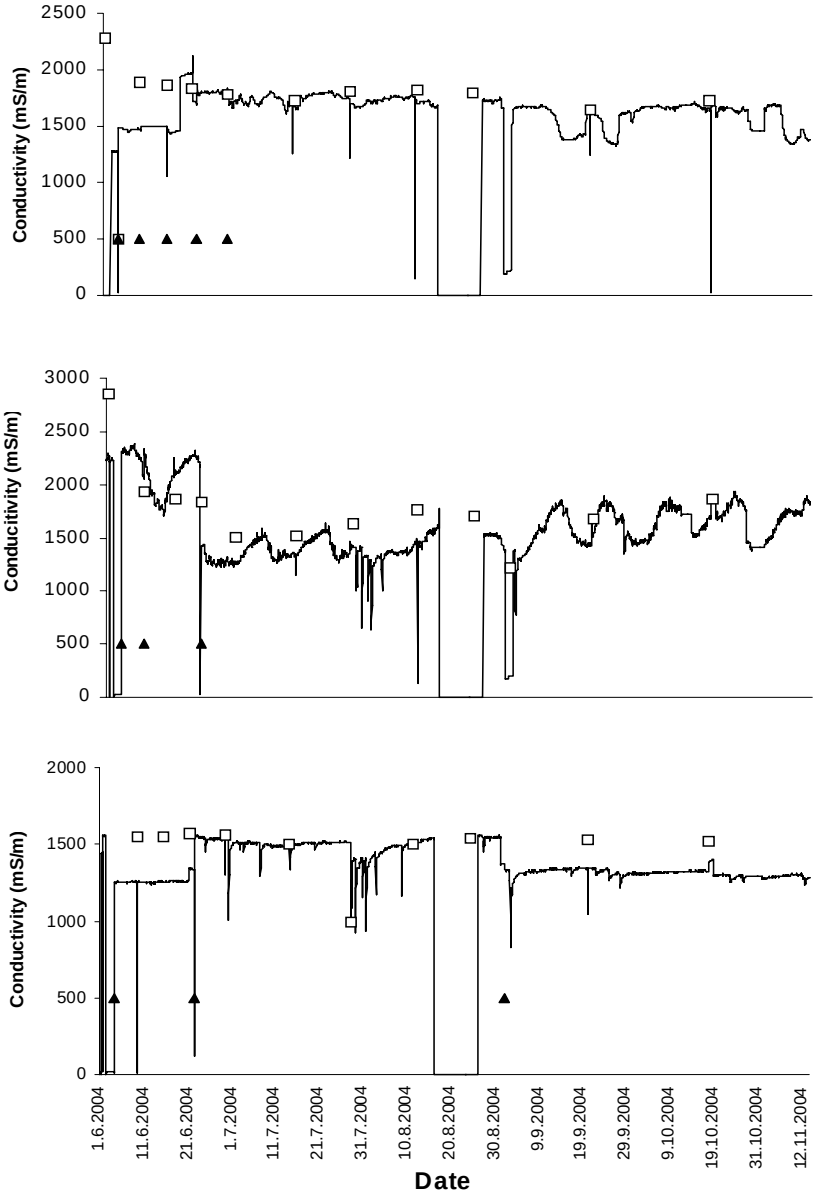

FIGURE 4 On-line electrical conductivity (--) in C1 (up), G3 (middle) and H1 (down), and calibrations (▲) and electrical conductivity of leachate samples measured by field meter (□).

3.3 Water table, temperature, conductivity and pH monitoring in Kujala

Water table and temperature were measured (on-line) in four (C3, G2, G3 and H1; 212-516 days) conductivity in three (C1, G3 and H1; 167 days) and pH in one monitoring well (H1; 167 days) in Kujala. The water table varied within approximately 8 meters between the four monitoring wells, while in the same monitoring wells the variation was mainly within one meter; for example, the water table increased slowly from 104 m to 105 m in G3 and H1 (Figure 5).

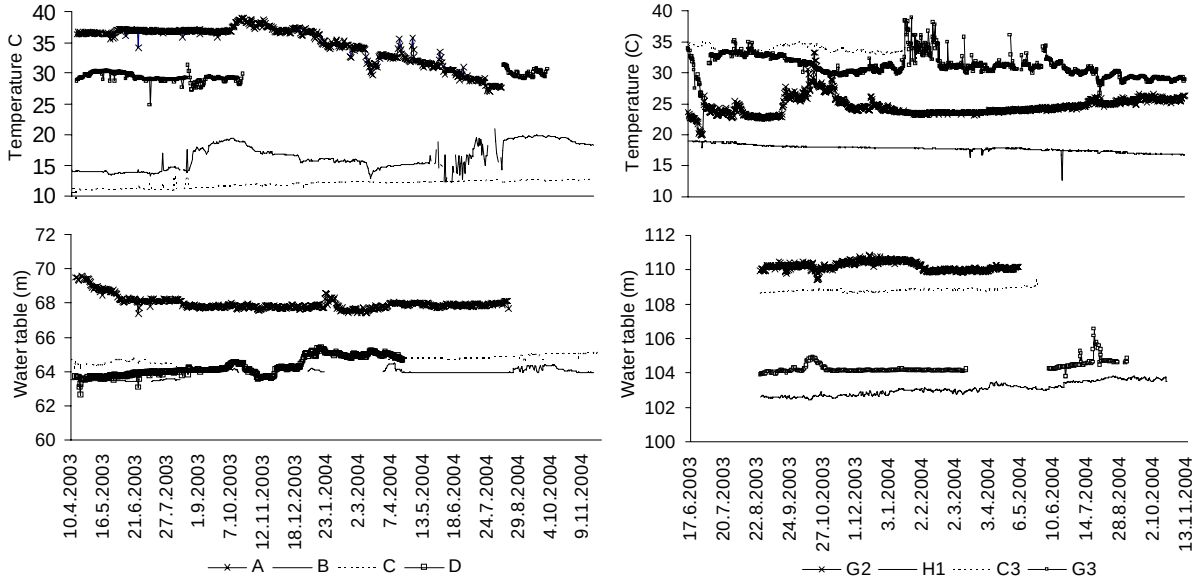

FIGURE 5 On-line measurements of temperature and water table in Ämmässuo (left) and Kujala (right) landfills.

Temperature varied from 17 to 35 ° C between the four monitoring wells and in same monitoring well from 2 to 10 °C. The highest temperature was recorded in C3 (range 33 - 34 °C), while the highest individual value (35 °C) was measured in G3, where the temperature however decreased gradually during the study to 29 °C. In other two monitoring wells the temperature was lower, increasing from 23 to 26 °C in G2, and decreasing from 19 to 17 °C in H1. The average conductivity was higher and varied more in G3 (1300-2300 mS/m) than in the other monitoring wells studied (1300-2000 mS/m, C1; 1300-1600 mS/m, H1; Fig. 4). The variation in G3 coincided with the recirculation of more dilute leachate (conductivity of 300-700 mS/m) in the recirculation canal located at five meters from G3 (Fig. 6). Only minor variation was observed in pH, which ranged from 6.9 to 7.3, without showing a clear trend, in H1 (Fig. 3).

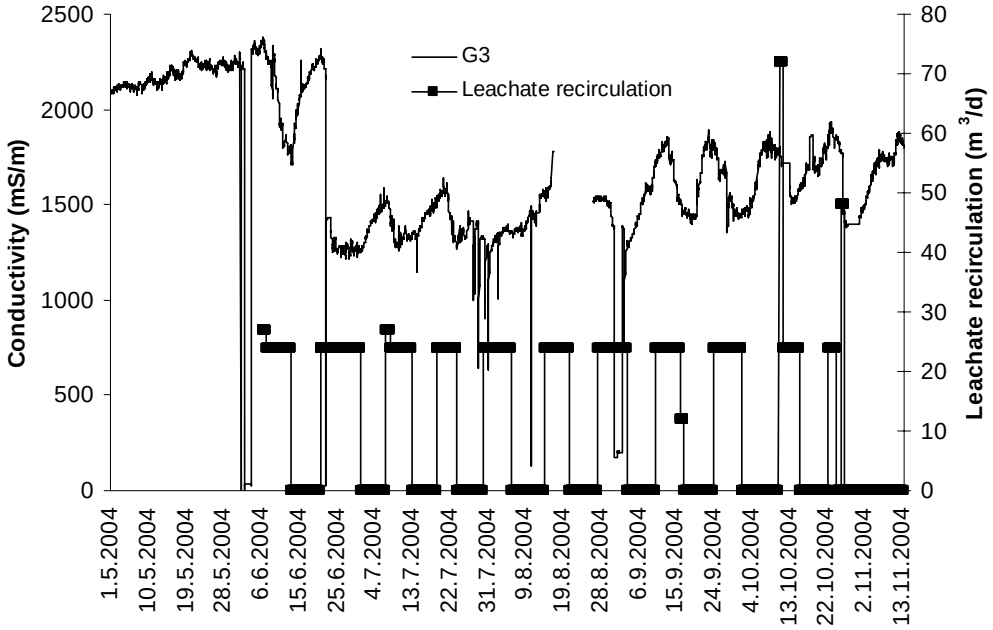

FIGURE 6 Electrical conductivity in leachate in the monitoring well (G3) during the period of leachate recirculation in 2004.

4 Discussion

In general, the on-line measurements indicated that the changes in the water table were low even during a monitoring period as long as one and a half year, while some gradual and temporary changes took place in temperature and electrical conductivity in some monitoring wells. Thus the usability of on-line measurements is highly case- and target- dependent. However, monitoring of water table and temperature has been considered important, especially when operating a landfill as a bioreactor, due to fact that rise in the water table may cause increasing leaching into groundwater and a high temperature can damage the lining, leachate and gas collection systems (Benson et al. 2007). Finnish legislation (Finnish Government 1997) requires monitoring of the water table in the internal leachate and the temperature within the landfill as well as the electrical conductivity in the external leachate. However, on-line monitoring is often not thought to be necessary, despite the fact that, as stated in the legislation (Finnish Government 1997), it is necessary to ensure that the various processes of degradation proceed as intended and that environmental protection structures (e.g., drainage layer, liner and leachate recirculation structures) are fully functional. Moreover it is important that monitoring is systematic, samples are representative and that changes in leachate quality are noted quickly. Thus the variation observed in electrical conductivity (present study) also demonstrated the difficulty of representative manual sampling as, for example, the concentration of ammonium nitrogen, which has been

generally considered one of the main pollutants in MSW leachate (Kjeldsen et al. 2002), has shown tendency to correlate with electrical conductivity (Marttinen et al. 1999).

This study shows the feasibility of on-line monitoring of electrical conductivity and pH in internal leachate in a MSW landfill. The values obtained from the on-line measurements were rather similar to those obtained by sampling and field measurements. The differences between the on-line and field meter values were systematic with respect to both parameters, the field meters tending to show higher values for electrical conductivity and pH than the on-line measurements. The lower values obtained from the on-line monitoring could be caused by landfill gas flow through leachate as landfill gas bubbles in leachate may decrease electrical conductivity, while release of carbon dioxide dissolved in leachate tends to increase the pH of samples immediately after sampling. The variation in the on-line measurements of electrical conductivity may indicate changes in internal leachate flow or characteristics as this variation coincided with the leachate recirculation in a monitoring well in Kujala where the electrical conductivity of the recirculated leachate was lower (range mainly 300 - 500 mS/m) than the internal leachate as a whole (mean approximately 1500 mS/m). However, a similar variation in electrical conductivity was not detected by field measurements performed approximately once in month; thus on-line monitoring seems to be a more accurate method of monitoring short-term variation in electrical conductivity.

Technically, the systems investigated in this study appeared to work without any major problems in both landfills. The on-line measurements worked well, and the data were collected through wireless data transfer, i.e., datalogger and data transfer by GSM modem to a computer with a photovoltaic energy supply. However the electrodes need regular maintenance, and further need for calibrations may exist, particularly in the longterm. It is difficult on the basis of the present study to generalize regarding minimum calibration intervals as it has previously been reported (Thomas & Pouet 2005) that the function of on-line measurements is highly dependent on wastewaters characteristics, e.g., the wastewater properties causing fouling of electrodes. It seems that maintenance (including cleaning and calibration) intervals between 1 and 2 months for electrical conductivity and pH might be appropriate in the landfill studied here. Contact between landfill gas and electronics should be avoided when implementing measurements, as a previous study (Kim et al. 2005) has shown that a major proportion of the sulphur in landfill gas exists in the form of hydrogen sulphide, which is highly corrosive. In addition to corrosion, settling of the landfill may cause some movement or bending in the monitoring well; thus monitoring wells with an inner diameter of >50 mm are highly recommended to prevent electrodes sticking and facilitate maintenance. Moreover foaming of the leachate in monitoring wells was seen in both studied landfills. This can cause inaccuracy in water table monitoring, and thus monitoring wells less prone to foaming are preferable for the purpose of monitoring.

In both landfills the highest temperatures (35 and 39 °C) approximated previously determined (reviewed by Barlaz et al. 1990) mesophilic range (34-42 °C) for optimal methane generation. It is clear that temperatures in a landfill are likely to vary in different parts and layers, as seen by the range from 5 to 39 °C previously measured in Ämmässuo (Sormunen et al. 2004). In warmer climates less vertical variation in temperature exists. Yazdani et al. (2006) reported variation between waste lifts of <10 °C in a landfill cell operated as a bioreactor and that the ambient temperature (range approximately 5-25 °C) had only a minor effect on temperatures. The present study suggests that variation in the temperature of the internal leachate is in part due to the height of landfill, at least in landfills located in a boreal climate. This is probably caused by the fact that the upper layers act as thermal isolation for the bottom layers. In fact inhibition of exothermic waste decomposition reactions may also happen in shallow landfill sections during winters. However, one monitoring well in Ämmässuo showed temperatures from 16 to 27 °C lower (height of landfill 20 m) than those in two other monitoring wells of similar height (17 and 24 m). This evidently show that besides the height of the landfill, waste quality and/or phase of degradation or some unknown reason, e.g., leachate flows, density and landfill geometry, can have effect on the temperature of the internal leachate.

In the present on-line measurements marked changes were not observed in internal leachate temperature in either landfill during the one and a half year period, despite the annual variation in ambient temperature of from -28 to 30 °C. Thus landfill gas production in the saturated zone of the landfills will probably continue without showing significant seasonal variation. In monitoring well G3 a decreasing trend from 35 to 29 °C was observed during a seven-month period in 2004, which might indicate a decrease in microbiological activity or changes in leachate flow around that area of the landfill. Variation in water table levels to leachate recirculation was not observed in present study. A previous study (Morris et al. 2003) found that leachate recirculation had minor effect on leachate generation, while leachate generation seemed to vary seasonally, a major effect on leachate generation being the infiltration of large amounts of rainwater. In the present landfills the water tables were generally rising and the saturated zone between the monitoring wells varied from 21 to 44 % and from 20 % to 51 % of landfill height in Ämmässuo and Kujala, respectively. A rising water table indicates that infiltration is higher than the accumulation of water into waste, as it has previously been found (Bengtson et al. 1994) that the accumulation of water continues even in 10-year-old landfills.

5 Conclusions

- On-line monitoring of the water table, temperature, electrical conductivity and pH of internal leachate is feasible in MSW landfills, and the measurement data can be collected using an automated system with wireless data transfer.
- Continuous monitoring of electrical conductivity may provide information of use in determining the representativeness of leachate sample, as local variation can occur, even in the same monitoring well. Thus the mean level of electrical conductivity could serve as and indicator of a representative leachate sample.
- On-line monitoring showed that variation in temperature and water table are generally slow and that marked seasonal variation does not occur in internal leachate in boreal landfills.
- In general on-line monitoring can be used to provide further information about conditions and changes in landfills. Its utility in traditionally operated landfills and landfills in which a small amount of leachate is recirculated is case-specific as changes in internal leachate are typically slow.

Acknowledgements

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IV

DETERMINATION OF FIRST ORDER KINETIC METHANE GENERATION FACTORS (RATE *k* AND POTENTIAL L_o) AND METHANE GENERATION POTENTIALS FOR TWO FINNISH MUNICIPAL SOLID WASTE LANDFILLS

by

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Submitted.

Determination of first order kinetic methane generation factors (rate k and L_o) and methane generation potentials for two Finnish municipal solid waste landfills

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Abstract

The aim of this study was to determine the methane generation factors (k) and methane generation potentials (L_o) for bulk waste in two Finnish landfills in order to provide information about methane potentials and emissions as well as the rate of degradation in landfills located in a boreal climate. The methane generation rates in the studied landfills, which were modelled (USEPA Landgem 3.02) with actual gas recovery rates and data obtained in the previous waste characterisation study, verified the k of 0.17 and L_o of 130 m^3/t found in Ämmässuo and k of 0.05 and L_o of 40 m^3/t found in Kujala for bulk waste. The modelling demonstrated that a major part of the methane emissions may take place before the implementation of gas collection system and that methane emissions may account for 56 % and 60 % of the methane generation in Ämmässuo and in Kujala, respectively, even if gas recoveries were implemented before landfills closure. Moreover the results demonstrated that carefully planned waste sampling may be a feasible way to determine k, which may be highly different between landfills even when located in same climatic conditions.

Key words: landfill gas, methane potential, emissions, recovery rate, modelling.

1 Introduction

Landfilling has been the main method of waste disposal during the last few decades, e.g., in the EU as well as in United States (Eurostat 2005, USEPA 2006). Waste streams of different kinds continue to end in landfills, with waste quality varying in according to the development and design of products, consumption and local waste management practices. Consequently, landfills are very heterogenous in their material composition. Even though source segregation of recyclable materials (paper and cardboard, metals, glass, biowaste) has increasingly been implemented since the 1990s in many countries, as in Finland, the proportion of biodegradable materials in landfilled household waste is likely to be high. For example, in Kujala and Ämmässuo it was 59 and 70 %, respectively (PHJ 2006, YTV 2004). In landfills biodegradable organic waste degrades in the course of time under anaerobic conditions, producing methane. The methane can be recovered and flared to reduce impact of the climate and/or used for energy production to replace fossil fuels. The proportion of recovered landfill biogas (of which approximately 50 % is methane) was 63 % of total biogas production in the EU (European Commission 2006). On the other hand methane emissions from landfills account approximately for 3 to 4 % of annual global (IPCC 2006) and approximately 3 % of Finnish annual anthropogenic greenhouse gas emissions (Statistics Finland 2006), while approximately 53 % of the total methane emissions in Finland originates from landfills (Statistics Finland 2006). It has been estimated that from 10 to 20 % of world anthropogenic methane emission originates from landfills (IPCC 2001).

Landfill gas generation models have been developed to estimate long-term gas generation rates in landfills. Several gas generation models have been developed (Reinhart et al. 2005, Scharff & Jacobs 2006) and most of these are based on first order reaction kinetics of biological degradation (e.g., USEPA Landgem, Formula 1; IPCC 2006). The reaction kinetics of landfilled waste have been estimated using the methane generation rate constant (k, year^{-1}), which is considered to be affected by several case-dependent factors, such as waste moisture, temperature of landfill, availability of nutrients and pH (USEPA 2005), and oxidation reduction potential, waste composition, waste density, alkalinity and particle size. Thus significant variation within the same landfill and between different landfills exists (Garg et al. 2006). It is generally accepted that precipitation, which affects the moisture of waste in landfill body, is the major factor determining methane generation, while the landfill temperature, biodegradability of waste and landfill depth are minor factors (Garg et al. 2006). In practice, k values are estimated on the basis of the recovered landfill gas flows and gas recovery efficiencies (Reinhart et al. 2005), gas emission measurements (e.g., flux chambers), and the local waste quality and landfilling practice. In addition to k the methane generation potential (L_o m^3/t) of the landfilled waste is necessary for modelling methane generation and is

estimated preferably on the basis of the waste composition or the measured methane generation potential of samples of the waste to be landfilled.

The reported k values for bulk waste in US landfills have varied widely from 0.003 to 0.21 year^{-1}, thus demonstrating the difficulty of determining or selecting site-specific k using the values in the literature (Garg et al. 2006). Landfill conditions are highly affected by local climatic conditions and by other local factors, e.g., waste management practices; thus using the same default values proposed in models or in the literature can be misleading, e.g., the regulations under the clean air act (CAA) suggest default k of 0.05 year^{-1} for conventional MSW landfills and in arid conditions the recommended k is 0.02 year^{-1} (USEPA 2005). Moreover a k of 0.04 for conventional landfills is provided on the basis of the emission factors in the AP-42 (Compilation of Air Pollutant Emission Factors, USEPA 2005). Recently k of 0.3 has been presented for well designed wet landfills, if the lag phase (acidogenic) is minimized (Faour et al. 2007). Although climatic conditions are the main factors determining k, the effect of a boreal climate, where a high proportion of the annual precipitation may be snow, on k have not been presented in the most recent studies, while new information on k in warmer climate conditions has been obtained in the latest bioreactor landfill field studies (Faour et al. 2007, Yazdani et al. 2006, Benson et al. 2007).

The default L_o (methane generation potential) given in a landfill gas generation model vary from 96 to 170 m^3/t waste (wet weight) (USEPA 2005). On the basis of recovered methane the L_o of 100 m^3/t might be appropriate in wet landfills (Faour et al. 2007) For a comparison Jokela et al. (2002) reported a biological methane potential (BMP, termed L_o in present study) of 130 m^3/t total solids (TS) for approximately ten-years-old landfilled MSW in Ämmässuo landfill (Espoo, Finland), while in a previous study L_o for unsorted fresh MSW ranged from 78 to 152 m^3/t TS (Barlaz et al. 1989). Source-segregated biowaste may have an L_o as high as 410 m^3/t TS while the residual MSW material (of which biowaste, paper and cardboard, glass and metals are source-segregated) may have L_o from 46 to 100 m^3/t TS (Jokela et al. 2002).

The objective of this study was to determine the kinetic factors (methane generation rate (k), methane potentials (L_o)) for two landfill a 20 (Ämmässuo) and 50 years (Kujala) old, respectively, landfills located in a boreal climate (Southern Finland) in order to estimate the methane generation potential and recovery rates in the those landfills. Moreover the emphasis was on the estimation of model parameters (k and L_o) for methane generation instead of the methane recovery as presented previously (Faour et al. 2007). Both landfills had significantly different histories and waste management practices. The k and L_o used were verified, mainly on the basis of a previous waste characterization study (Sormunen et al. 2008) and gas recovery data from particular landfills.

2 Materials and methods

2.1 Modelling methane generation

The methane generation rates in the two landfills were modelled using the Landgem 3.02, landfill gas emission model (equation 1, USEPA 2005). Besides annual landfilling rates the default k and $L_\circ$ constants provided with the Landgem model (k varying from 0.02 to 0.7 and $L_\circ$ varying from 96 to 170 m^3/t depending on either values based on inventories or determined in the Clean Air Act (CAA) and landfill location in wet or arid areas) were used as well as k and $L_\circ$ determined on the basis of previous experimental studies (Sormunen et al. 2008) of waste samples from the studied landfill bodies. Moreover gas generation rates modelled using the different k and $L_\circ$ values were compared to the actual volumes of methane recovered through the gas recovery systems in the two landfills.

$$Q_{CH4} = \sum_{i=1}^{n} \sum_{j=0,1}^{1} kL_\circ \left(\frac{M_i}{10} \right) e^{-kt_{ij}} \qquad (1)$$

Where:
Q_{CH4} = annual methane generation in the year of calculation (m^3/a), i = 1 year time increment, n = (year of calculation) - (initial year of waste acceptance), j = 0.1 year time increment, k = methane generation rate ($year^{-1}$), $L_\circ$ = methane generation potential (m^3/t), M_i = mass of waste accepted in the i^{th} year (t) and t_{ij} = age of the j^{th} section of waste mass M_i accepted in the i^{th} year (decimal years).

2.2 The studied landfills

The study was performed using data from two MSW landfills (Ämmässuo and Kujala), which contain approximately 9 (Ämmässuo) and 3.1 (Kujala) million tons of MSW and have been operated for approximately 20 and 50 years, respectively as previously described by Sormunen et al. (2008). Ämmässuo is the largest Scandinavian landfill, and accepts waste from the metropolitan area of Finland (approximately 1 million inhabitants), while Kujala receives waste from a smaller city (Lahti, approximately 100 000 inhabitants) landfill. In the 1990s annual landfilling rates ranged from 0.4 to 0.6 * 10^6 tonnes in Ämmässuo and from 50 to 80 * 10^3 tonnes in Kujala. The waste history and the operation of the two landfills were highly different between the two landfills. In Ämmässuo, since 1987 all the MSW has been landfilled as such and source segregation increasingly implemented, the aim being to achieve only residual waste landfilling by the 1990s.

In contrast, in the early phases of Kujala landfill MSW was landfilled as incineration ash as the incineration of MSW was practised from 1965 to 1984 in the Lahti region. Also in Kujala, on-site burning of waste was generally practised from the 1960s to 1980s, sludges were landfilled until 1980, and

leachate recirculation was practised during the summer from the 1970s to the 1990s. Thus on the basis of their landfill histories Ämmässuo landfill contains relatively more biodegradable waste than Kujala landfill. The most recent (YTV 2004, PHJ 2006) waste characterisation studies have shown that the biodegradable fraction of the present household waste may still account up 70 % in Ämmässuo and up to 60 % in Kujala, and thus considerable amount of biodegradable waste has been landfilled in recent years, despite source segregation of biowaste and paper as well as cardboard in residential buildings. Waste incineration is not practised in either region, except for some co-combustion of recycled fuels (REF), which may partly explain the high proportions of biodegradable compounds in the landfilled waste. Landfill gas recovery systems (and energy utilisation) have increasingly been implemented in Ämmässuo since 1996, while in Kujala a gas recovery system (including 20 vertical wells and 6 horizontal trenches) was implemented in 2002; no additional gas recovery wells have been introduced since then. The vertical gas recovery wells were installed in an area, which had reached its final height before 2002, while the horizontal gas recovery trenches were used in areas still in the filling stage. The landfill is to be closed in the end of the 2007 in accordance with the EU landfill directive (1999/31/EC), which requires sealed bottom structures for MSW landfills.

In the present study the methane recovery rates (m^3/a) were determined by summarising the reported monthly gas flows and methane concentrations at the gas recovery plants. In Ämmässuo annual methane recovery increased from 3.2 to 37.4 million m^3/a (approximate methane content 52 %) during 1996-2006. During this period approximately 200 gas recovery wells and four pumping stations were built. In Kujala methane recovery rose from 1.4 to 1.8 million m^3/a (methane content 43-45 %) in 2002-2004. At that time temporary cover layers (e.g., surplus soils) were increasingly used to decrease gas emissions and increase the gas recovery rate. The data on the volumes of extracted gas and amounts of landfilled waste were obtained from the landfill operators.

2.3 Determinations of k and L_0 for bulk waste

Previously the Ämmässuo and Kujala landfills were characterized using landfill body waste samples taken at different horizontal locations and from different heights of the landfills (Sormunen et al. 2008). The characterization study took into account various characteristics, e.g., TS, volatile solids (VS) and L_0 of waste samples (Sormunen et al. 2008). The experimentally determined L_0s were approximately 110 and 30 m^3/t (wet weight) in the top layers in Ämmässuo and Kujala, respectively. In addition to these values, methane generation was modelled using higher L_0s (130, 150 and 170 m^3/t for Ämmässuo and 40, 50 and 60 m^3/t for Kujala) on the grounds that the contemporary L_0s were probably higher than those in the 2- to 3-year-old samples used in our previous study (Sormunen et al. 2008). The ks based on the waste characterisation study (Sormunen et al. 2008) were determined on the basis of the half-life time ($t_{1/2}$) of

L_o, assuming that it corresponds to the mass-based half-life time of degradable organic carbon and using the previously reported (IPCC 2006) relationship between k and $t_{1/2}$ for degradable organic carbon: $k=ln(2)/t_{1/2}$. For Ämmässuo half-life was calculated using the linear trend between the L_o of waste landfilled in 2003 (130 m^3/t, in the upper layer of landfill sector 1) and L_o of waste landfilled around 1987 (average 10 m^3/t in the bottom layer of sector 3). For Kujala the half-life of L_o was calculated on the basis of the L_o in the upper layer (landfilled in 1998-2003) and L_o in one of the bottom layers (landfilled in 1984-1989) located approximately 7 to 9 meters from the bottom structure. Moreover a k similar or close to (0.05, 0.06, 0.07 for Ämmässuo and 0.03, 0.04, 0.05, 0.06 for Kujala) the k of 0.04 previously determined for conventional landfills (USEPA 2005) was screened in the present study for both landfills with previously determined L_os of 110 and 30 m^3/t.

3 Results

3.1 Determination of k on the basis of the experimentally determined L_o

The k for the two landfills was determined on the basis of the half-life time ($t_{1/2}$) of L_os. In Ämmässuo $t_{1/2}$ would be approximately four years and thus k 0.17 $year^{-1}$ (Table 1), while in Kujala $t_{1/2}$ would be approximately 14 years and k 0.05 $year^{-1}$.

TABLE 1 Determination of k on the basis of L_o (Sormunen et al. 2008) and their half lives in Ämmässuo and Kujala landfills.

Ämmässuo						
Years of landfilling	n	Mean TS (%)	Mean VS (%, dry weight)	Mean L_o (m^3/t)	Half-life (½) of L_o (years)	k ($k=\ln(2)/t_{1/2}$)
2001-2002 (sector 1, top layer)	3	61	71	113	4	0.17
1987-1992 (sector 3, bottom layer)	6	50	55	11		
Kujala						
1998-2003	3	67	51	30		
1984-1989	2	67	36	15	14	0.05

3.2 Evaluation of methane generation and recovery rates by varying k and L_o

Methane generation in the studied landfills was modeled using different k and compared to the amounts of methane recovered in those landfills (Fig. 1).

Modeling was performed using the experimentally obtained k of 0.17 for Ämmässuo and 0.05 Kujala as well as the values of 0.05, 0.06, 0.07 for Ämmässuo and 0.03, 0.04, 0.06 for Kujala with experimentally obtained L_o of 110 m^3/t for Ämmässuo and 30 m^3/t for Kujala (Table 2). The present methane generation varied from 31 to 50 $*10^6$ m^3/year in 2006 and from 1.7 to 2.2 $*10^6$ m^3/year in 2005 in Ämmässuo and Kujala, respectively, depending on the k used (Fig. 1). From 2007 onwards (both landfills will be closed in 2007) the methane generation seems likely to decrease faster rate in Ämmässuo than Kujala due to higher k; e.g., with k of 0.17 methane production from 2007 to 2008 will fall by approximately $9 * 10^6$ m^3/a in Ämmässuo.

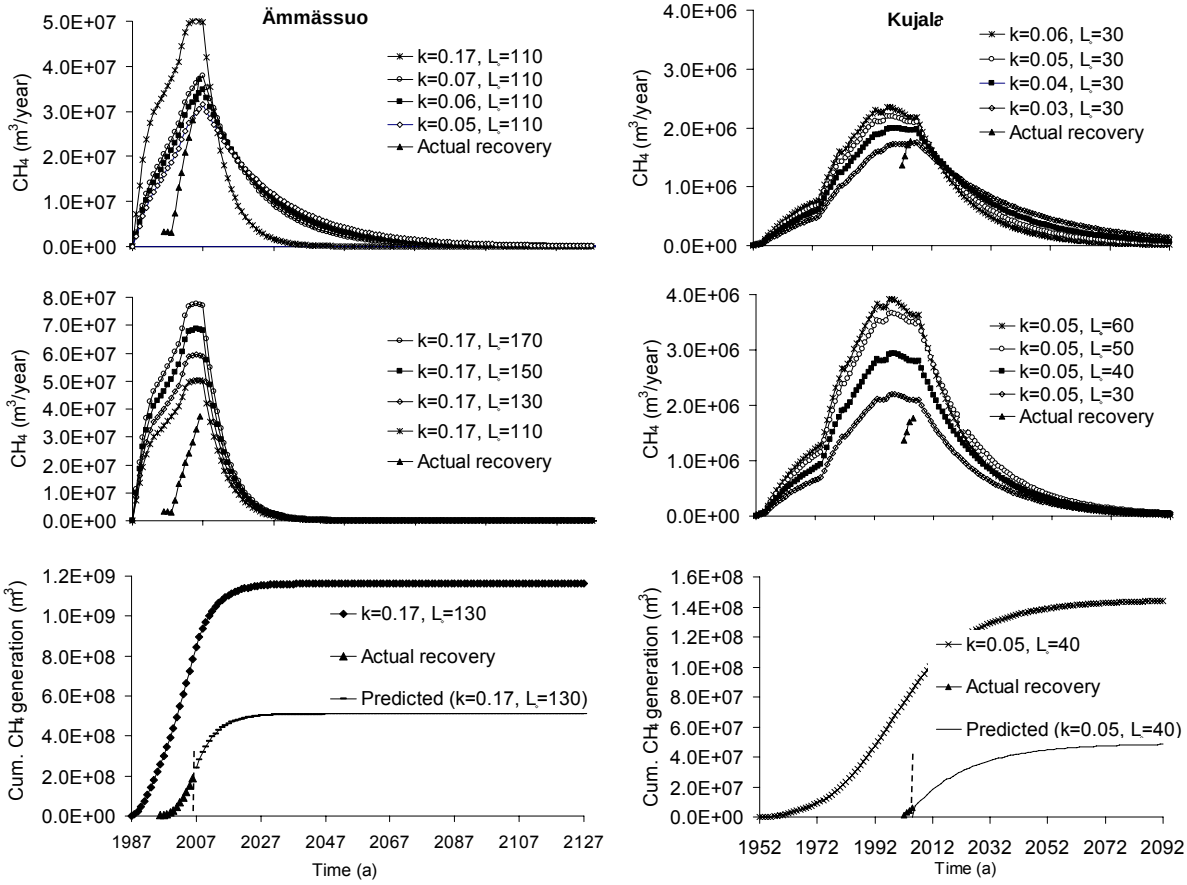

FIGURE 1 Methane production by varying k (top) and varying L_o (middle) compared to actual methane recovery and the modelled cumulative methane generation (bottom) by experimentally determined k and L_o in Ämmässuo and Kujala.

The reliabilities of k and L_o for the two landfills were estimated by the proportion of methane recovery calculated according to the methane generation modelled with the varying k and L_o values. However methane recovery was increasingly implemented and optimized by gas characteristics (CH_4 and O_2 concentrations), gas flow and suction pressure at the gas revovery plant in Ämmässuo between 1996 and 2004 and optimized (as in Ämmässuo) between 2002 and 2005 in Kujala, which increased the methane recovery efficiencies in

both landfills. For example with k of 0.05 and L_o of 110 m^3/t methane recovery efficiency would have increased from 20 % (in 1996) to 122 % (in 2006), whereas with k of 0.07 and L_o of 110 m^3/t methane recovery increased from 16 % (in 1996) to 101 % (in 2006) and with the previously (in this study) determined k (0.17) and L_o (110 m^3/t) methane recovery increased from 9 % (in 1996) to 75 % (in 2006) in Ämmässuo (Table 2).

TABLE 2 The methane recovery rates (% of modeled methane production) as modelled by the Landgem (USEPA) model with different k and L_o in Kujala and Ämmässuo landfills.

Ämmässuo	L_o = 110 m^3/t, varying k from 0.05 to 0.17				k = 0.17, varying L_o from 130 to 170 m^3/t		
Year	L_o = 110 k = 0.05	L_o = 110 k = 0.06	L_o = 110 k =0.07	L_o = 110 k = 0.17	L_o = 130 k = 0.17	L_o = 150 k = 0.17	L_o = 170 k = 0.17
1996	20	17	16	9	8	7	6
1997	19	16	15	9	8	7	6
1998	16	14	13	8	7	6	5
1999	36	32	29	19	16	14	12
2000	61	54	49	32	27	24	21
2001	69	61	56	37	31	27	24
2002	82	72	66	44	37	32	28
2003	88	78	71	48	41	36	31
2004	99	88	80	56	48	41	36
2005	110	99	91	65	55	48	42
2006	122	110	101	75	63	55	48

Kujala	L_o = 30 m^3/t, varying k from 0.03 to 0.06				k=0.05, varying L_o from 40 to 60 m^3/t		
	L_o = 30 k = 0.03	L_o = 30 k = 0.04	L_o = 30 k = 0.05	L_o = 30 k = 0.06	L_o = 40 k = 0.05	L_o = 50 k = 0.05	L_o = 60 k = 0.05
2002	79	69	64	61	50	38	36
2003	88	77	71	68	54	43	41
2004	99	87	81	78	61	49	47
2005	102	91	85	81	63	51	49

In Kujala gas recovery efficiency with k of 0.03 and L_o of 30 m^3/t increased from 79 % (2002) to 102 % (2005), whereas with the k of 0.06 and L_o of 30 m^3/t methane recovery efficiency increased from 61 % (in 2002) to 81 % (in 2005) and with the present determined k of 0.05 and L_o of 30 m^3/t recovery efficiency increased from 64 % (in 2002) to 85 % (in 2005) in Kujala.

The methane generation rates were also studied using higher L_o (from 130 to 170 m^3/t and from 40 to 60 m^3/t in Ämmässuo and Kujala, respectively) than determined in the waste characterisation study (Sormunen et al. 2008), with the experimentally determined k (of 0.17 for Ämmässuo and 0.05 for Kujala) (Table 2). The methane recovery efficiencies in Ämmässuo then varied from 8 % (in 1996) to 63 % (in 2006) with L_o of 130 m^3/t, while with L_o of 170 m^3/t the methane recovery efficiencies varied from 6 % to 48 %. In Kujala methane recovery efficiencies varied from 50 % (in 2002) to 63 % (in 2005) with L_o of 40 m^3/t, while

with higher L_o (60 m^3/t) the methane recovery rates varied from 36 % (in 2002) to 49 % (in 2005).

3.3 Total methane generation potential

The total methane generation potential of the two landfills during their lifetime was modeled using experimentally determined L_o (110 m^3/t for Ämmässuo and 30 m^3/t for Kujala) and also higher L_o (130 m^3/t for Ämmässuo and 40 m^3/t for Kujala), as some degradation was evident prior to sampling for the experimental analysis of L_o in both landfills. The modeled total methane generation varied from 984 (L_o of 110 m^3/t) to 1 163 * 10^6 m^3 (L_o of 130 m^3/t) and from 108 (L_o of 30 m^3/t) to 144 * 10^6 (L_o of 40 m^3/t) in Ämmässuo and Kujala, respectively (Table 3). k has no effect on the total methane generation, but the time needed to achieve a certain proportion of the total methane generation potential is determined by k. Thus the k determined in the present study by the experimental data (0.17 for Ämmässuo and 0.05 for Kujala) were used in the models for both landfills. The results showed that, e.g., approximately 90 % of the total methane generation potential will be achieved in Ämmässuo in 2013 (k of 0.17) and in Kujala in 2020 (k of 0.05). Assuming that 85 % of the generated methane will be recovered the remaining (from 2006 onwards) recoverable methane potentials are 319 *10^6 m^3 (L_o of 110 m^3/t) and 378 *10^6 m^3 (L_o of 130 m^3/t) with k of 0.17 in Ämmässuo and 45 *10^6 m^3 (L_o of 30 m^3/t) and 59 *10^6 m^3 (L_o of 40 m^3/t) with k of 0.05 in Kujala. Thus during the lifetime of the landfills the proportion of recovered methane will vary from 47 % (L_o of 110 m^3/t) to 44 % (130 m^3/t) with k of 0.17 and from 33 (L_o of 30 m^3/t) to 40 % (40 m^3/t) with k of 0.05 of the total methane potential in Ämmässuo and Kujala, respectively (Table 3, Figure 1).

3.4 Methane emissions

The proportion of methane emissions was calculated by subtracting the proportion of the total recovery potential (461 and 510 *10^6 m^3 in Ämmässuo; 43 and 48 *10^6 m^3 in Kujala) from the total methane generation potential (984 and 1 163 *10^6 m^3 in Ämmässuo; 108 and 144 *10^6 m^3 in Kujala) (Table 3). The total methane emission potentials are 523 *10^6 (L_o of 110 m^3/t) and 653 *10^6 m^3 (L_o of 130 m^3/t) in Ämmässuo as well as 65 *10^6 (L_o of 30 m^3/t) and 96 *10^6 m^3 (L_o of 40 m^3/t) in Kujala. The remaining (from 2006 onwards) methane emission potentials are 48 *10^6 (L_o of 110 m^3/t) and 57 *10^6 (L_o of 130 m^3/t) with k of 0.17 in Ämmässuo, while in Kujala the remaining methane emissions are 7 * 10^6 m^3 (L_o of 30 m^3/t) and 9 *10^6 (L_o of 40 m^3/t) with k of 0.05, if 85 % of the generated methane is recovered in both landfills. The proportions of methane emissions before the implementation of methane recovery were 19 % (L_o of 110 m^3/t and 130 m^3/t) and 41 % (L_o of 30 m^3/t and 40 m^3/t) of the total methane generation potentials in Ämmässuo and Kujala, respectively. The major (81 %) proportion of the methane emissions was generated before the implementation of the gas

recovery system in Kujala, while the corresponding proportion was minor (32-34 %) in Ämmässuo.

TABLE 3 The total and remaining methane potentials as well as recovery potentials by varying L_o (and k) of Ämmässuo and Kujala landfills.

	Ämmässuo (*10^6)		**Kujala** (*10^6)	
Actual cumulative methane recovery	190 (from 1996 to 2006)		6 (from 2002 to 2005)	
	L_o = 110 m^3/t and k = 0.17	L_o = 130 m^3/t and k = 0.17	L_o = 30 m^3/t and k = 0.05	L_o = 40 m^3/t and k = 0.05
Total methane generation potential	984	1 163	108	144
Total recovery potential[a]	461	510	43	48
Total methane emission	523	653	65	96
Methane emissions before implementation of gas recovery[b]	185	219	55	73
Remaining methane generation potential since 2006 (Ämmässuo) and 2005 (Kujala)	319	378	45	59
Remaining methane emissions[c]	48	57	7	9

[a]Actual recovery + modeled methane generation by recovery rate of 85 % of the generated methane since 2006 in Ämmässuo and since 2005 in Kujala, [b]Gas recovery implemented in 1996 in Ämmässuo and in 2002 in Kujala, [c]From 2006 onwards in Ämmässuo and from 2005 in Kujala, assuming that 85 % of the generated methane is recovered (methane oxidation not considered).

4 Discussion

The methane generation factors (k) and methane potentials (L_o) of MSW are critical factors in modeling gas generation rates for landfills as the results can be used, e.g., for planning gas recovery and upgrading and utilisation (heat, power, traffic fuel) systems as well as for the inventory and trading of greenhouse gas emissions. These factors are also used by IPCC (IPCC 2006). The present results demonstrated that the k and L_o in the studied two Finnish landfills are apparently different, even if the landfills are located only at

approximately 100 km distance from each other and the climatic conditions (e.g., mean annual precipitation 601-700 mm) are very similar in both cases.

The L_o of landfilled waste (as bulk waste until the 1990s and within the last decade increasingly as a source-segregated grey waste) seems to be approximately 130 m^3/t (wet) in Ämmässuo and approximately 40 m^3/t (wet) in Kujala. These, present, values are suggested by the experimental determination of L_o and comparison of the modelled methane generation with the amounts of methane actually recovered in the last 2-3 years, when the gas recovery system has been fully implemented and both landfills are close to their final heights. The actual original methane generation potentials of the landfilled waste was probably higher than the L_o experimentally determined from the waste samples taken from the upper layers of the landfill bodies, in which waste had been landfilled within the 2-3 years before sampling in both landfills. This, is likely to be the case as it has been assumed (e.g., IPCC 2006) that methane production is highest in the first few years of landfilling due to degradation of easily degradable materials. On the other hand easily degradable materials may induce the acidogenic phase of degradation, when methane production is limited, as a lag of approximately 2 years in methane production was considered typical in US landfills, and even in so called wet landfills where moisture content is specified by a bioreactor landfill operation (Faour et al. 2007).

The wide discrepancy in L_o between the two landfills studied here is probably caused by difference in the effectiveness of the source segregation of biodegradable waste (e.g., biowaste, paper and cardboard) in these two regions. In both regions biodegradable waste has been source- segregated since the 1990s, but in the Lahti (Kujala) region source segregation is more effectively performed than in Ämmässuo; for example the proportion of kitchen biowaste and soft tissue together were 28 % (YTV 2004) and 23 % (PHJ 2006) of the landfilled waste in Ämmässuo and Kujala, respectively. Moreover energy waste and landfilled waste have been source-segregated in all households, and in residential buildings containing more than 10 households landfill waste, energy waste, biowaste, paper and cardboard are source-segregated in the Lahti region, while in the metropolitan region source segregation have not been implemented in buildings with less than five households and in residential buildings with more than 10 households only paper and biowaste have been source segregated (YTV 2004). The content (% of total waste mass) of organic matter (measured as VS of dry weight) was higher in Ämmässuo than in Kujala, which may indicate also the existence of higher amounts of degradable organic matter in Ämmässuo (Sormunen et al. 2008). Moreover, the content of TS was lower in Ämmässuo, which means that the moisture content was more favorable to biodegradation in Ämmässuo than in Kujala. On the other hand the longer stabilization time with leachate recirculation in Kujala probably caused a higher stage of stabilization, especially in the bottom parts of the landfill, which may have caused a decrease in VS and low methane generation following the implementation of methane recovery. The proposed L_o (130 m^3/t)

for bulk waste for Ämmässuo is within the range proposed by the US EPA landgem model (170 m^3/t, laid out by the Clean Air Act, CAA, or 96 and 100 m^3/t based on the EPA's Compilation of Air Pollutant Emission Factors, AP42), while the L_o for bulk waste in Kujala is lower than the default (100 and 170 m^3/t for conventional landfills) values. In Kujala the proposed L_o (40 m^3/t (wet) was low compared to the biodegradable (e.g., garden waste, wood, papers and cardboard, textiles, residual biowaste, nappies) fraction, which still accounted for up to 59 % of the landfilled household waste in a previous waste characterisation study (PHJ 2006). The fact that in the modeling the experimentally determined low L_o fitted the actual amount of recovered methane could be due the fact that the landfill also contains inert materials, e.g., surplus soils, industrial waste and incineration ash deposited between 1965 and 1984 (Sormunen et al. 2008). Moreover on-site burning of waste was also practised in Kujala. On the other hand the L_o used for landfilled waste in Kujala (30 m^3/t wet and 44 m^3/t TS) was similar to that previously determined for source-segregated grey waste (46 m^3/t TS, Jokela et al. 2002). This in turn, may indicate a slow rate of degradation in grey waste during the first two-three years of landfilling, as in a previous study (batch assays with added inoculum, Jokela et al. 2002) the k determined for grey waste was 0.03 during an incubation period of 237 days.

The experimentally determined k values (k of 0.17 for Ämmässuo and 0.05 for Kujala) indicated a much higher rate of degradation in Ämmässuo than in Kujala. This was probably caused by the higher proportion of easily degradable waste, such as kitchen biowaste, as it has been found that the biodegradable fraction can account for as much as 70 % (of which 25.2 % may consist of kitchen biowaste) of the landfilled household waste in Ämmässuo (YTV 2004), where as in Kujala the corresponding figure was 60 % (PHJ 2006). For comparison a default k of 0.06 (range 0.05-0.08) is used in national greenhouse gas inventories (IPCC 2006) for the degradation of rapidly degrading waste such as food waste in dry temperate conditions (conditions determined by the ratio of mean annual precipitation/potential evapotranspiration, MAP/PET<1), while in wet temperate conditions (MAP/PET>1) approximately the same k (range 0.1-0.2, default 0.185) is used for food waste as that proposed in the present study (0.17) for landfilled bulk waste in Ämmässuo. On the other hand the present k of 0.17 for methane generation in Ämmässuo was lower than the estimated k of 0.3 by Faour et al. (2007) for recovered methane in wet (specified moisture by leachate recirculation and moisture addition) landfill conditions. The US EPA defines a bioreactor landfill according to a moisture content of 45 % (wet weight) in landfills where leachate has been used for moisture addition (Reinhart et al. 2005). Previously (Faour et al. 2007) k was determined for methane recovery, whereas present to study determined k for methane generation, and thus it seems that a k of 0.17 may be appropriate for Ämmässuo. Moreover the high moisture (mean moisture content of 46 %, Sormunen et al. 2008) in Ämmässuo may favor biodegradation as higher k (from 0.3 to 0.5) has been reported for wet landfills in previous studies (Faour et

al. 2007, Yazdani et al. 2006). In Kujala the determined k of 0.05 is same as the default k (range 0.04-0.06) used by IPCC (2006) for moderately degrading waste such as garden waste in dry temperate conditions (MAP/PET<1). The fact that the present k of 0.05 determined in Kujala is much lower than that in Ämmässuo is probably caused by the fact that a larger part of the landfill body was more stabilized during the several decades of landfill's operation, and thus the present k for bulk waste is low, although it is evident that with newer waste k will be higher.

In the present study the first order kinetic factors (k and L_o) for methane generation were determined on the basis of the experimentally determined L_o values in the two landfills and their feasibility was assessed/confirmed by comparison of the modeled methane generation with the recovered methane. This methodology could provide for more accurate modeling of the methane production potential of landfills than modeling based on the values for actually recovered methane. In most previous studies models and methane generation factors (k, L_o) have been validated using recovered methane (m^3 methane) and assumptions about gas extraction efficiency, methane oxidation and methane emission. In reality these factors assumed factors are generally not known due to fact that they are very case- dependent (mainly moisture and temperature) and whole site methane emission measurements have been rarely used in methane generation models (Scharff & Jacobs 2006).

The present and previous studies (Laurila et al. 2005, Spokas et al. 2006) suggest that approximately two thirds of the methane produced is recovered in landfills with temporary covers. In Ämmässuo 63 % of the generated methane was recovered in 2006 (based on proposed k of 0.17 and L_o of 130 m^3/t) and a similar recovery rate was obtained in Kujala with proposed lower k (0.05) and L_o (40 m^3/t). The recovery rates were approximately the same as those previously determined by methane recovery data and micrometeorogical emissions methods for three different sections (mean 62 %, range 44-72 %) of Ämmässuo landfill in 2003 (Laurila et al. 2005) and previously suggested (65 %) for temporarily covered landfills for the European Pollutant Emission Register (EPER) as a part of the IPCC (Spokas et al. 2006). However, as methane production will apparently continue in a landfill for several decades, recovery efficiency can be improved through the implementation of additional gas recovery wells and final (less impermeable) cover layers. In landfill cells with clay and geomembrane final covers recovery efficiencies from 84 % to 98 % have been reported (Spokas et al. 2006).

The present results on modeling methane generation in landfills indicate the importance of the implementation of a gas recovery system in determining the fate of the methane generated. In the two landfills studied here approximately 56 % (Ämmässuo) and 60 % (Kujala) of total methane generation during the landfills' history will be released without treatment, partly due to fact that approximately 19 % and 41 % of the total methane generation occurred before the implementation of gas collection systems in Ämmässuo (1996) and in Kujala (2002), respectively. Improved gas recovery efficiency (from 8 to 63 % in

1996-2006) has been obtained in Ämmässuo due to fact that the gas recovery has been implemented in larger areas and some areas have been covered by final cover structures. Part of the methane which is not recovered is biologically oxidized to carbon dioxide in the landfill cover layers; e.g., a methane oxidation rate of 30 % was observed in temporary cover layers such as surplus soils and compost materials in Ämmässuo (Laurila et al. 2005) and a methane oxidation rate of 4 to 50 % for a variety of cover designs with a vertical or horizontal gas recovery system have been determined (Spokas et al. 2006). Apparently oxidation can be optimized in temporary and final cover layers through the optimisation of factors (surface methane flow, temperature, water content and nutrients) important for methane oxidation (Kettunen et al. 2006, Einola et al. 2008). Thus the final methane emissions from landfills are affected, not only by managing the waste composition also through the proper timing and intensity of the methane recovery system, as well as through methane oxidation.

5 Conclusions

- The determination of the biological methane generation potentials of waste samples with carefully planned sampling seems to be feasible way to determine first order kinetic factors (k and L_0) for bulk waste.
- The appropriate k for bulk waste were 0.17 in Ämmässuo and 0.05 in Kujala, which indicates a higher rate of degradation in Ämmässuo than proposed by the default k of 0.09 (IPCC 2006) for a wet and temperate climate, while in Kujala the rate of degradation seems to be the same as the proposed default k of 0.05 (IPCC 2006) for bulk waste in landfills located in dry and temperate conditions.
- The L_0 of 130 m^3/t and 40 m^3/t were determined for bulk municipal solid waste in Ämmässuo and Kujala, respectively, thereby indicating high differences in local waste management and landfilling practices.
- Methane emission accounted for 19 % and 41 % of the total methane generation potentials before the implementation of gas recovery systems in Ämmässuo and Kujala, respectively.
- Methane emissions into the atmosphere may account for 56 % and 60 % of the total methane potentials in Ämmässuo and Kujala, respectively.

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LEACHATE AND GASEOUS EMISSIONS FROM INITIAL PHASES OF LANDFILLING MECHANICALLY AND MECHANICALLY-BIOLOGICALLY TREATED MUNICIPAL SOLID WASTE RESIDUALS

by

Kai Sormunen, Juha Einola, Matti Ettala & Jukka Rintala 2007

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Leachate and gaseous emissions from initial phases of landfilling mechanically and mechanically–biologically treated municipal solid waste residuals

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Abstract

In this study, the behaviour, and leachate and gaseous emissions during the initial phases of landfilling mechanically (M) and mechanically–biologically (MB) treated municipal solid waste residuals in northern climatic conditions was compared using two landfill lysimeters (112 m^3). The results demonstrate that the strong acid phase of M residuals degradation lasts at least 2 years, while in the MB residuals the acid phase lasts only a few months. The SCOD and NH_4–N concentrations varied 20–100 g/l and 600–1800 mg/l in M leachate and 1–4 g/l and 100–400 mg/l in MB leachate, respectively. The leaching of SCOD was approximately 40-fold (24.2 and 0.6 kg/t TS) and leaching of NH_4–N approximately 5-fold (356 and 60 g/t TS) from the M than MB residuals; thus the effect of biological stabilisation was more marked on the leaching of SCOD than of NH_4–N. Moreover gas (methane, carbon dioxide and nitrous oxide) emissions were several-fold higher from the M than MB residuals.

Keywords: Mechanical–biological; Nitrogen; Organic matter; Methane; Landfill

1. Introduction

Landfilling has been the main means of disposal of the source-segregated grey waste fraction of municipal solid waste (MSW) during the last few decades. However, in the European Union new legislation has been introduced for reducing the amount of landfilled waste and environmental pollution caused by landfills, thus emphasis is on avoidance of waste generation, and material re-use or energy utilization of waste. According to EC directive (1999/31/EC) share of biodegradable landfilled MSW has to be reduced by 25% before the year 2006, compared to the amount of biodegradable waste in 1994 and, further, by at least 50% before 2009 and 65% before 2016. In some EU countries even more demanding national requirements were set on the basis of the EC directive so that in Germany, for example, it has been possible to landfill only thermally and mechanically–biologically pre-treated MSW since June 2005 (Stegmann, 2005). Source segregation of biowaste (kitchen and garden waste), papers, cardboard and energy waste (e.g., plastics and non-recyclable papers such as tissues, paper cups, food containers, brown papers and binders) might be enough in some EU countries to meet the aims for 2006, while the further requirements (from 2009 onwards) needs other methods such as mechanical, mechanical–biological treatment or incineration to meet the target for reduce remaining share of biodegradable landfilled MSW.

The aim of the mechanical treatment of MSW is to separate organic materials for use as recycled fuel (REF), metals for reuse and to prepare the residual waste for further treatment. Mechanical treatment employs, e.g., shredding,

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sieving and other sorting methods, which means that the mechanically produced residuals (M residuals) have smaller particle size and the waste is mixed. When this type of residuals is landfilled the initial phase of degradation may be enhanced (as compared to non-processed waste) or this in turn, may inhibit methane production due to the increased production of organic acids (reviewed by Komilis et al., 1999). Smaller particles may also enhance leaching of organic materials and nitrogen (Bone et al., 2003). However M residuals are usually post-treated biologically by aerobic or anaerobic processes or by a combination of these, which significantly reduces the gaseous and leachate loads from landfills (Stegmann, 2005). The leaching potentials of organic compounds and nitrogen from mechanically–biologically treated residuals (MB residuals) have been studied in several laboratory studies (e.g., Cappai et al., 2005; Höring et al., 1999; Leikam and Stegmann, 1997), while landfill lysimeter scale studies, especially in cold climatic conditions, have been rather neglected. To our knowledge comparative studies of landfilling M and MB residuals with measured leachate and gaseous emission as well as conditions in waste body have not been reported.

The objective of this study was to evaluate gaseous and leachate emissions from the initial phases of the landfilling of M and MB residuals. For this study two 112 m^3 landfill lysimeters were constructed, and filled with separately produced M and MB residuals. The landfill body conditions (temperature, pore gas), gaseous emissions (flows of CH_4, N_2O, CO_2) as well as leachate flow and characteristics (pH, conductivity, organic material, nitrogen) were followed systematically over a period of about 2 years and the emissions were related to the initial characteristics of the residuals.

2. Methods

2.1. Wastes

M residual was obtained from Loimi-Häme Regional Solid Waste Management Ltd. (Forssa, Finland). In the Loimi-Häme region metals are source-segregated, and biowaste and papers are source-segregated in residential buildings containing more than five households, while in the case of other buildings biowaste, paper, cardboard and glass are source-segregated where the segregated waste streams are >20 kg per week. Furthermore, a network of local collection points for papers, metals and batteries exists for households. The residual and grey waste fraction is further processed in a mechanical plant. The processing includes pre-shredding, screening, removal of non-magnetic and magnetic metals, shredding, other magnetic removal and drum screening (50 mm). The fraction >50 mm is processed to produce REF, while the sieved (<50 mm) fraction was used in this study as the M residual. The M residual was transported in trucks to the landfill site operated by Mustankorkea Ltd. (Jyväskylä, Finland) and was stored for 2–3 days outdoors before landfilling into lysimeters.

The MB residual was prepared by composting of M residual (described above) in seven batches for 2–3 weeks in aerated pilot tunnels (two 50 m^3 tunnels, Vapo Biotech Ltd., Jyväskylä) followed by passively aerated pile composting outdoors for 6–14 months. Wood chips (0.5 m^3/t M residuals) or the composted oversize fraction (>15 mm, 0.5 m^3/t M residuals) of the MB residual was used as support material in the tunnel compost. The compost was mixed by a front loader weekly during the tunnel composting and two-three times in the first 2–3 months of the pile composting. The gas generation within 21 days of testing (GB21 value) was 22 Nl/kg TS after 3 weeks' tunnel and 5 weeks' pile composting as determined from one of the seven batches (Lehtinen, 2003). This would have been close to the requirements presented in the German landfill ordinance (AbfAblV, 2001), which requires that mechanical biological treatment should stabilize waste so that its gas production potential over a period of 21 days measured by a standardised method (GB21) should be <20 Nl/kg TS. Finally, all the composted MB residuals were mixed together and screened in the drum (40 mm) to remove the support materials.

For the analyses and determinations the M residual sample (60 l) was prepared by combining six 10 l samples (obtained one from each truck container), while the MB residual sample was combined from five 10 l samples obtained from randomly selected locations in the mixed MB residual pile. For the nitrogen analyses approximately 1 l of M and MB residual from the above-mentioned mixed 60 and 50 l samples was homogenised into particles below 2 mm by a cutting mill (Retsch SM2000). The characteristics of the M and MB residuals are shown in Table 1.

2.2. Lysimeters

The two landfill lysimeters (height 3.9 m, width 2.4 m, length 12 m, volume 112 m^3) made from steel frames (RHS 60 × 80 mm) and walls (2 mm) and coated with acryl paint (Hempatex Hi-build 46410) were placed in a 30 years old waste and soil landfill body in November 2003 (Fig. 1).

Table 1
The characteristics of M and MB residuals in the landfill lysimeters

Parameter	M	MB
Wet weight (t)	84	97.8
TS (%) and (t TS)	67 and 56.3	54 and 52.8
VS[a] (%) and (t VS)	41 and 34.4 t	23 and 22.5
VS/TS (%)	61	43
pH	5.9–6.5	7.0–7.6
BMP (m^3/t TS) and (m^3/t wet)	259 and 105	52 and 21
N_{tot} (%) and (kg)	0.5 and 412	0.6 and 548
Height (m)	3.2	3.5
Volume (m^3)	92	96
Density (t/m^3)	0.9	1.0 (cover layer 0.8)[b]

[a] VS of wet weight.

[b] The cover layer contained gravel 10–15 cm and 40–45 cm (9.8 t, slightly compacted) MB residual.

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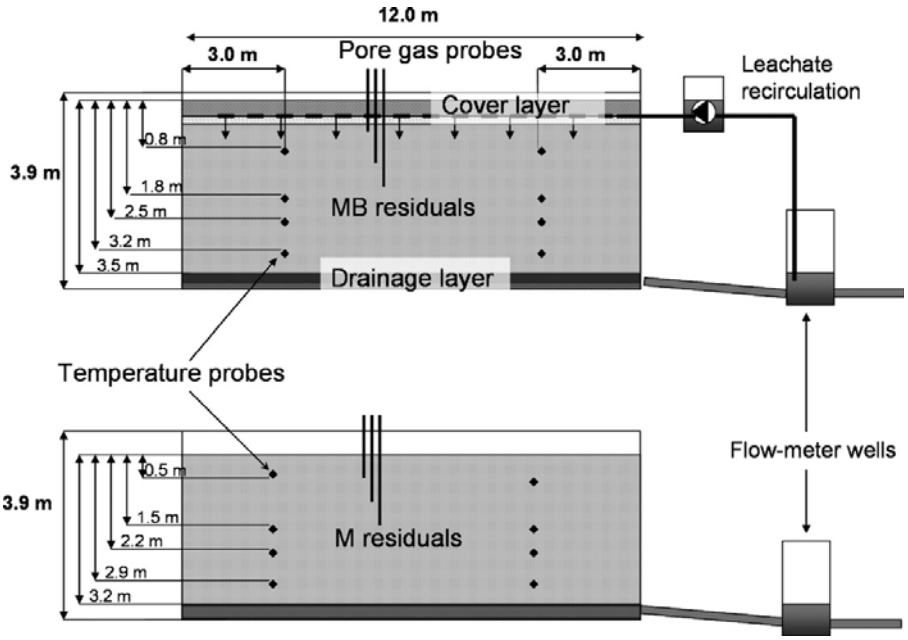

Fig. 1. Sketch of MB (top) and M residual (bottom) lysimeters placed in an old landfill body.

The lysimeters were filled with the M and the MB residuals in 0.5 m horizontal layers, which were compacted by a soil compactor (Bomag 105, 1.6 t). The densities obtained were 0.9 and 1.0 t/m^3 in M and MB residuals, respectively (Table 1). The leachates were gravimetrically (angle 5%) collected from the drainage layer (thickness 30 cm, gravel particle size <25 mm) and collection drain (110 mm) to flow-meter wells. The lysimeters were covered by a plywood board covers during approximately the first 120 days (from 1st of December 2003 to 1st of April 2004) and thus the leachate flows were low before April 2004. In June 2004 tap water (2 × 500 l) were added to the lysimeters to promote leachate formation and to monitor the leachate flow by a tracer (lithium chloride) method (data not shown). The added water (1000 l) was counted as extra rainfall.

During a specific study period (140 days, from 8th of June till 24th of October 2005) the leachate from the MB residual flow meter well was recirculated back in the MB lysimeter in order the study the effects of leachate recirculation on gas production and leachate characteristics. The leachate was recirculated through a distribution layer (thickness 150 mm, gravel particle size <25 mm) located below 40–45 cm (slightly compacted) cover layer of MB residual. The system of distribution pipes consisted of a 40 m pipe (Ø 25/31 mm, PVC), of which the last 10 m was perforated (Ø 5 mm holes, interval between holes 0.5–1 m). The leachate flow (water table in the well) was continuously measured by a pressure meter (Keller PR-36W) and a datalogger (Campbell Scientific CR10X).

The temperatures within the landfilled M and MB residuals were monitored by a soil temperature and moisture station (Davis 6343) and temperature probes (Davis 6470) with a wireless Vantage Pro console (Davis 6310). Two parallel series (four probes per series) of temperature probes were located at two sites in both lysimeters (Fig. 1) and the results for each depth were reported as mean values of two parallel probes. Ambient air temperature was monitored by a weather station (Davis Vantage Pro 6150 equipped with a datalogger/PC-link 6510) on the landfill area. Rainfall data were obtained from the Finnish Metereological Institute (2006).

2.3. Analyses and determinations

Total solids (TS), volatile solids (VS), soluble chemical oxygen demand (SCOD), ammonium–nitrogen (NH_4–N) and Kjeldahl nitrogen (N_{tot}) were analysed as previously described by Sormunen et al. (2007). Biological methane potential (BMP) was determined with digested mesophilic municipal sewage sludge as an inoculum (Nenäinniemi Jyväskylä, Finland) in triplicate 2 l glass vessels as previously described by Sormunen et al. (2007). Biogas volume was measured using a displacement method. Methane and volatile fatty acids (VFA) were measured by a Perkin–Elmer Autosystem XL gas chromatograph with flame-ionization detector as described in Luostarinen and Rintala (2005).

pH and redox were measured by a WPA (CD70) meter and Sensorex pH 450 CD and Sensorex ORP 450 electrodes, and conductivity by a Hanna instruments (Hi 9635) conductivity meter. Pore gases (CH_4, CO_2 and O_2) were measured by an IR analysator (Geotechnical Instru-

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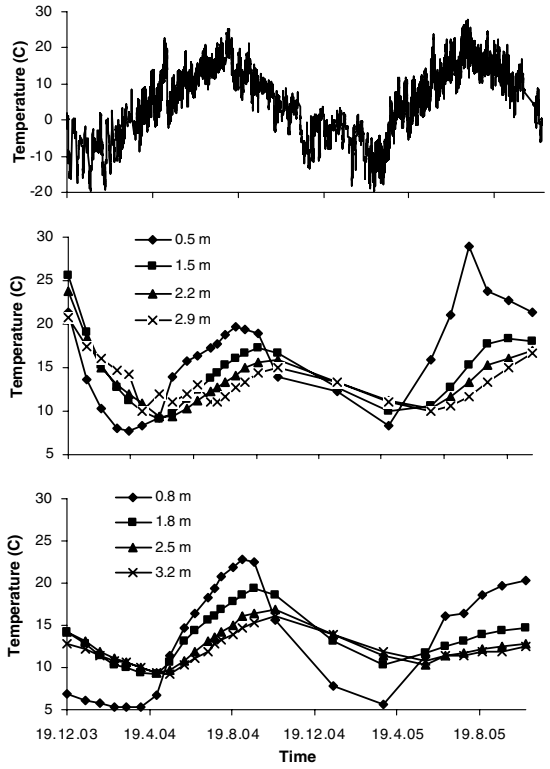

Fig. 2. Ambient temperature (top) and temperature in M (middle) as well as in MB residual (bottom) lysimeters at four different depths. Depths were measured from top of the waste body.

ments GA 94) with steel tubes (Ø 30 mm) at depths of 0.5, 1.0 and 1.5 m. Gas emissions (CH_4, CO_2, N_2O) were measured by a FTIR analysator (Gasmet DX4000) at five sampling points (distance about 2 m) with a flow chamber (Ø 50 cm and volume 29 l) method.

3. Results

During the study period, the temperatures ranged from 8 to 29 °C and from 5 to 23 °C in the M and MB lysimeters, respectively (Fig. 2), while the ambient temperature ranged from −19 to 27 °C. The highest (21–26 °C) temperatures were detected at the beginning of the landfilling (December 2003) in the M residual, after which temperatures remained at approximately the same level in both residuals until towards the end of the study period (June 2005 onwards), when the temperature in the M increased more than in

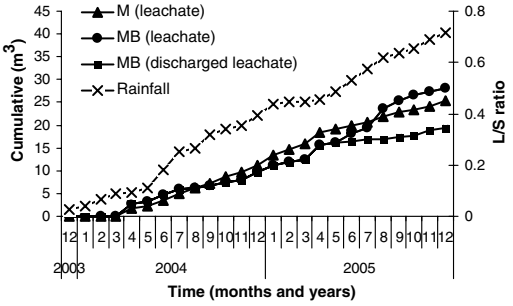

Fig. 4. Cumulative leachate volumes and leachate/solids (L/S) ratios of M and MB residuals and rainfall in 2004–2005. The discharged leachate differs from leachate volume in MB residuals, due to leachate recirculation.

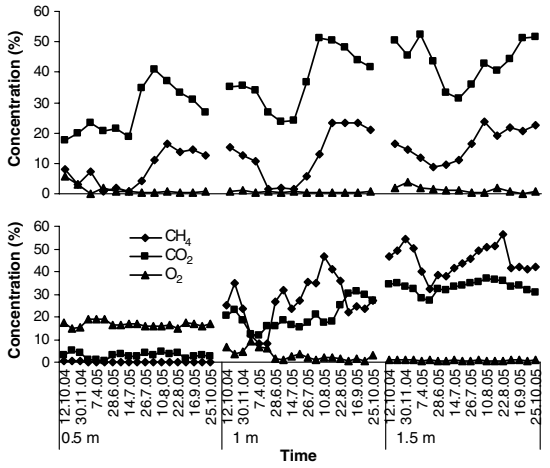

Fig. 3. Pore gas concentrations at different depths of M (top) and MB residuals (bottom).

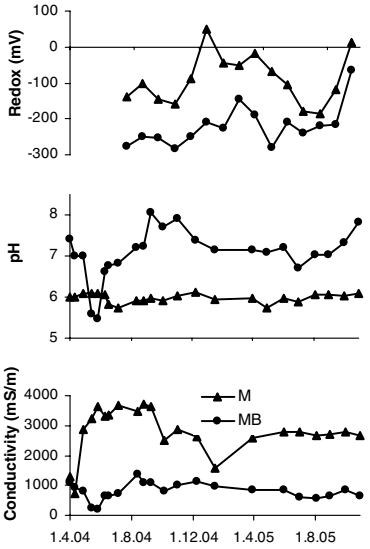

Fig. 5. Redox-potential (top), pH (middle) and conductivity (bottom) of M and MB leachates.

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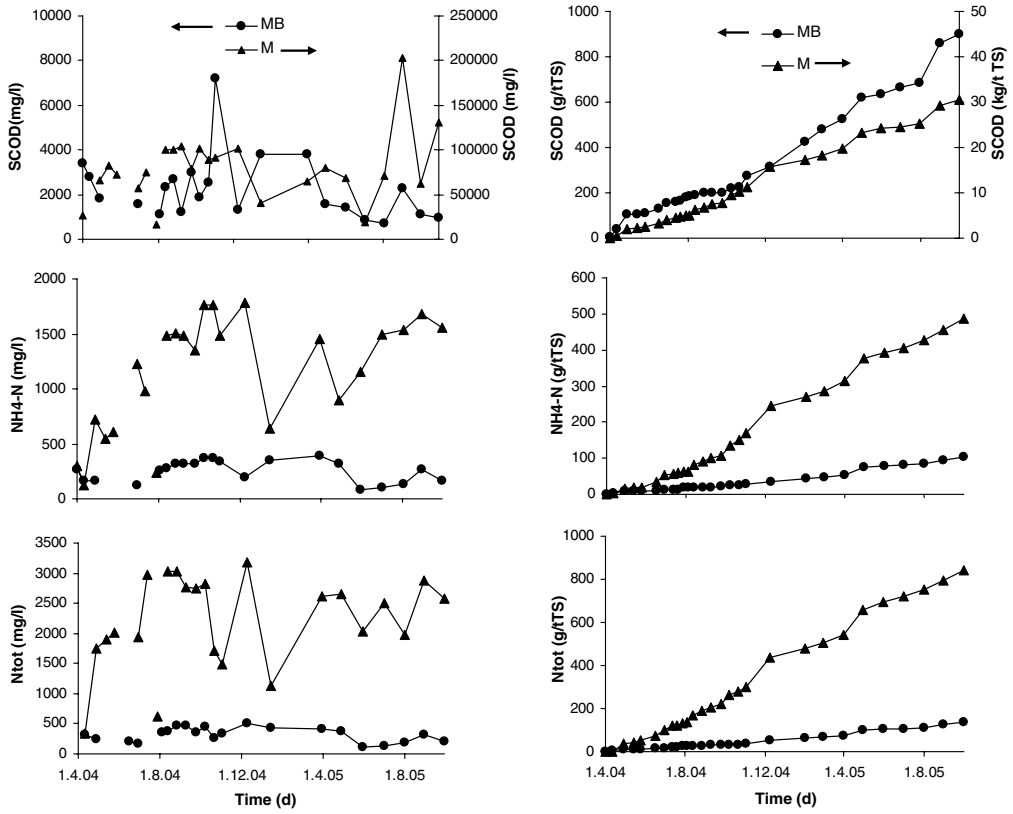

Fig. 6. SCOD (top), NH_4–N (middle) and total N (bottom) concentrations (mg/l) and leachate loads (g/t TS or kg/t TS) of M and MB residuals. Leachate recirculation was practised during 140 days, from 8th of June till 24th of October 2005.

the MB lysimeter. In both lysimeters, the temperature in the highest stratum (0.5–0.8 m) corresponded most clearly to the ambient temperature. During the summers of 2004 and 2005 (from June to October) the top layer of the MB landfill was totally covered by vegetation, while the M landfill remained almost free of vegetation.

Among the pore gases in the landfill body O_2 was detected mainly at the depth of 0.5 m in MB lysimeter and also at 1.0 m in the MB lysimeter and 0.5 m in M lysimeter (Fig. 3). The CH_4 concentrations were higher in the MB than M lysimeter, except in the latter part of the study at depth of 0.5 m, while the CO_2 concentration was mostly higher in the M than MB lysimeter. After 1 year of landfilling (October 2004) the CH_4 and CO_2 concentrations at depth of 1.5 m were 47–54% and 33–34% in MB lysimeter, while in the M lysimeter the respective values were 12–16% and 46–53%.

The leachate flow increased when the snow started to melt due to increasing ambient temperatures in April 2004 (Fig. 4). During the study the highest daily leachate flows were about 0.7 m^3 and 0.9 m^3 (5.4.2005), while the monthly flows varied from 0.6 to 2.4 m^3 and from 0.4 to 3.2 m^3 in the M and MB lysimeters, respectively (data not shown). The cumulative leachate flows during the 426 d period before leachate recirculation in the MB lysimeter were 19.2 and 16.2 m^3 from M and MB lysimeters, respectively, corresponding to 70% and 59% of the cumulative rainfall (27.3 m^3). During the leachate recirculation period (140 d from June to October 2005) 8.9 m^3 (309 mm as precipitation, weekly 12–37 mm) leachate was recirculated back to the MB landfill body and 1.3 m^3 leachate was discharged from the MB lysimeter, while from the M residual the discharged flow during the same period was 3.4 m^3. At the end of the study (640 d) total cumulative leachate flows to the outside of the lysimeters were 25.3 m^3 and 19.4 m^3 (63% and 48% of rainfall) from M and MB lysimeters, respectively. The obtained annual leachate flow/solid (L/S ratios, in 2004) in the landfill bodies were 0.24 and 0.20 in M and MB, respectively, without leachate recirculation, while leachate recirculation increased the annual L/S-ratio in the MB body to 0.30 in 2005.

The pH of the M leachate varied from 5.8 to 6.1 during the study, while the pH of the MB leachate decreased from 7 to 5.5 after 1 month of landfilling for the following 2 weeks (Fig. 5). Afterwards pH of the MB leachate

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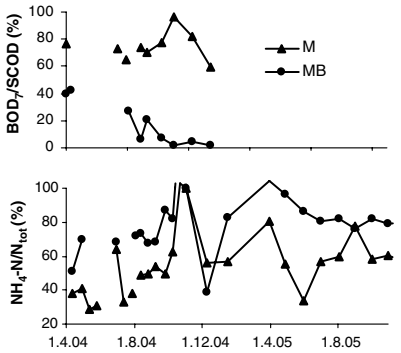

Fig. 7. BOD_7/SCOD- (top) and NH_4–N/N_{tot}-ratios (bottom) of M and MB leachates.

Table 2
The cumulative leachate loads of NH_4–N, N_{tot} and SCOD during the 426 day period before the leachate recirculation in the MB residual

	NH_4–N (g/t TS) (% of initial content)	N_{tot} (g/t TS) (% of initial content)	SCOD (kg/t TS)
M	391 (7.8)	694 (13.8)	24.2
MB	79 (1.3)	103 (1.7)	0.6

Leached proportions of NH_4–N and N_{tot} from initial nitrogen content of the M and MB residuals are shown in parenthesis.

increased to 6.8, and remained thereafter at 7–8 with increasing values after the summer season (September–October). The redox were throughout higher in the M (−200 to 0 mV) than MB leachate (−300 to −200 mV) and increasing values were observed after August in 2004 and 2005, and occasionally values above 0 mV in the M leachate. The M leachate had constantly higher conductivity (approximately 3000 mS/m) than the MB leachate (approximately 1000 mS/m). However conductivity showed a decreasing trend in both leachates during the study period.

NH_4–N (filtered samples) concentrations were 5–10-fold higher in the M (approximately 600–1800 mg/l, mean 1162 mg/l) than MB leachate (approximately 100–400 mg/l, mean 258 mg/l), where lower concentrations were found towards the end of the study period (Fig. 6). In both leachates, the concentrations of NH_4–N and its proportion of nitrogen content (NH_4–N/N_{tot} ratio, N_{tot} non-filtered samples) increased during the beginning of the study (until November 2004), more in the M leachate, thereafter the concentrations and NH_4–N/N_{tot} ratios varied without clear trends (Fig. 7). The NH_4–N/N_{tot} ratios were, excluding some individual samples, 40–70% and 70–90% in the M and MB leachate, respectively. During the period (426 d) before leachate recirculation the leaching of NH_4–N was about 5-fold more from the M (391 g/t TS) than MB residual (79 g/t TS), and the leaching of total nitrogen about 7-fold more from the M (694 g/t TS) than MB residual (103 g/t TS) (Table 2).

In the M leachate the SCOD values ranged from ca 20 to 100 g/l peaking at 200 g/l while in the MB leachate the SCOD values were initially typically 2–4 g/l levelling down to 1–2 g/l towards the end of the study period (Fig. 6), although the SCOD load in the MB leachate increased slightly at the end of study. During the period (426 d) before the leachate recirculation (in MB residual) approximately 40 times more SCOD was leached from the M (24.2 kg/t TS) than MB residual (0.6 kg/t TS).

BOD_7 and VFA in the leachates were determined for selected samples in 2004 and BOD_7 once in both leachates in 2005. In the M leachate the BOD_7/SCOD- (Fig. 7) and the VFA_{cod}/SCOD-ratios (Fig. 8) were 59–96% and 19–91%, respectively, while in the MB leachate the ratios decreased from the initial ~40% to <7% for BOD/SCOD in 6 months and from the initial ~48% to under 6% for

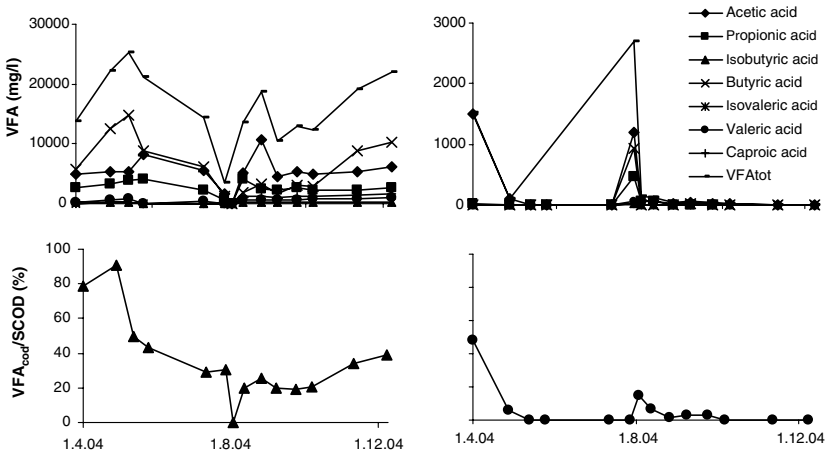

Fig. 8. VFAs (top) and VFA_{cod}/SCOD ratios (bottom) in M (left) and MB (right) leachates.

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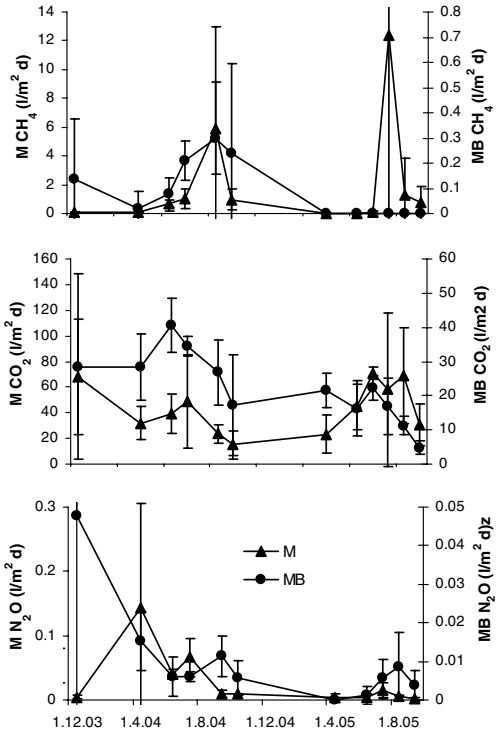

Fig. 9. The CH_4 (top), CO_2 (middle) and N_2O (bottom) emissions of M and MB residuals.

VFA_{cod}/SCOD in a month and remaining thereafter at <1% and peaking up to 15% after 3 months. The BOD values were 24 g/l in the M and about 0.05 g/l in the MB leachate after 10 months' (January 2005) leachate flow, while the respective SCOD values were 41 g/l and 3.8 g/l. The total VFA concentrations in the M leachate varied from 3.5 to 25 g/l and in the MB leachate from 0.009 to 2.7 g/l.

Carbon dioxide formed the major proportion of the gas emissions, ranging from 15 to 71 l/m^2 d (mean of separate measurements 43.7 l/m^2 d) and from 5 to 41 l/m^2 d (mean 22.4 l/m^2 d) in the case of the M and MB residuals, respectively (Fig. 9). Both methane and nitrous oxide emissions were higher from the M than MB residuals (range from 0.05 to 12.4 l CH_4/m^2 d (mean 1.9 l CH_4/m^2 d) and 0.001–0.14 l N_2O/m^2 d (mean 0.025 l N_2O/m^2 d) in M; range from <0.02 to 0.3 l CH_4/m^2 d (mean 0.08 l CH_4/m^2 d) and <0.001 to 0.05 l N_2O/m^2 d (mean 0.01 l N_2O/m^2 d) in MB).

4. Discussion

The results of the present comparative lysimeter study – which to our knowledge is one of the first on this scale, and especially in northern climatic conditions – clearly show that aerobic stabilization causes a marked reduction in gaseous and leachate emissions of mechanically processed municipal solid waste during the initial phase (25 months) of landfilling. The impact could be seen, e.g., in the 5-fold higher BMP, 5–10-fold higher SCOD values and 3–6-fold higher nitrogen concentrations in the M than MB leachate (similar leachate flows) and in the much smaller leachate loads from the MB than M residual, which were about 3% in SCOD, 20% in NH_4–N and 15% in nitrogen. Furthermore, the greenhouse gas emissions, namely methane and nitrous oxide were up to 20-fold and 11-fold higher, respectively, from the M than MB residuals, despite the fact that the M residual was still mainly in the acidogenic phase. It is clear that the SCOD and nitrogen concentrations and loads from the initial phases of the landfilling of MB residuals are much lower than in the case of M residuals, and as decreasing trends were not observed it appears that the loads continue approximately at the same rates for a while (except SCOD from MB-residual) from both materials after the period studied here.

These differences in leaching of SCOD and nitrogen are suggesting the need for different types of leachate treatment concepts and technologies. On the basis of MB residual landfill simulation study the NH_4–N and organic matter pollution may require treatment as long as L/S ratio of 3 and 2.5 will be reached, respectively, which may take in landfill conditions 150–200 years depending on discharge limits (Höring et al., 1999). Moreover some specific compounds which are seldom measured in leachates, e.g., phthalates and PAHs as well as toxicity may necessitate leachate treatment (Marttinen et al., 2003). The present SCOD values (mainly from 710 to 3827 mg/l) in MB leachate were a little higher or at the same level as in a laboratory study (mainly from 500 to 2000 mg/l determined as COD, Leikam and Stegmann, 1997) and in a landfill lysimeter study (COD from 762 to 3043 mg/l, Felske et al., 2003) or in a full-scale landfill study where the major part of the waste consisted of MB residuals (COD from 228 to 4670 mg/l, Bone et al., 2003). The present SCOD values in the M leachate varied from 16,000 to 203,000 mg/l and the values did not decrease during the study, which indicates a huge leachate pollution potential as the landfill conditions seem to remain strongly acidic (pH about 6) for at least 2 years. In practise some proportion of the SCOD could be degraded in methanogenic part of full-scale landfill, thus the leachate load of SCOD would probably be smaller in full-scale landfill than in present landfill lysimeter, where waste was in same age and conditions were more favourable for acidogenesis than methanogenesis. The proportion of VFA_{cod} from SCOD was 19–91% in the M leachate and mainly <1–15% in MB leachate, indicating that proportion of easily degradable organic material is high in M leachate. The NH_4–N/N_{tot} ratio increased in the MB leachate, indicating higher ammonification after 6 months of leachate flow, while the NH_4–N/N_{tot} ratio was about 20% lower in the M leachate. The nitrogen concentrations in the present study in the M leachate were about two thirds of the M leachate nitrogen concentration

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observed in other field study (Woelders and Oonk, 1999) and about the same as in a laboratory study (Leikam and Stegmann, 1997). The SCOD and NH_4–N concentrations in the MB leachate were slightly higher and N concentrations about same in the present study as in the corresponding laboratory study (Leikam and Stegmann, 1997) in the first 1–3 months of the landfilling, while later, during the following three to 5 months, the NH_4–N and nitrogen concentrations increased in both studies.

The present lysimeter studies as well as previous experiences from laboratory studies (e.g., Cappai et al., 2005; Leikam and Stegmann, 1997) and field studies show that M (Woelders and Oonk, 1999) and MB residual landfills (Bone et al., 2003; Felske et al., 2003) have different mechanical and biological characteristics compared to each other as well as to untreated MSW. Because of the higher biological stability of MB, temperatures are lower in MB than M landfills and conventional MSW landfills. In present study, temperatures were higher in M than MB residuals in the beginning of the landfilling and in the end of the study indicating higher biological reactivity in the M than MB residual. The densities of the waste bodies in this study were 0.9 t/m^3 and 1.0 t/m^3, while a density of 1.3 t/m^3 for a compacted M residual landfill (Woelders and Oonk, 1999), densities of 1.0–1.6 t/m^3 for MB residual landfills (e.g., Felske et al., 2003; Scheelhaase and Bidlingmaier, 1997; Stegmann et al., 2005) and densities of 0.5–0.8 t/m^3 typical for untreated waste landfills (Reinhart and Townsend, 1998) have been reported. Due to higher density the water permeabilities are lower in MB landfills (10^{-5}–10^{-10} m/s, Stegmann et al., 2005) than in untreated waste landfills (10^{-4}–10^{-8} m/s, Reinhart and Townsend, 1998), which may cause practical difficulties, e.g., reduced mechanical stability of the landfill body due to high pore water pressure (Stegmann et al., 2005). Moreover leachate recirculation may require a pressurised system when high recirculation rates (e.g., >30 mm per week) are used, as in the case of M residual (density 1.3 t/m^3) bioreactor landfill study (Woelders and Oonk, 1999). In the present study, high leachate recirculation rates (up to 33–37 mm/week) were used into MB lysimeter (1.0 t/m^3), thus showing that lower compaction may enable leachate recirculation by a gravity-based system. However, increased gas production was not observed due to leachate recirculation even though in the previous field-scale study (Lorber et al., 2001) the gas production was increased due to water addition in the MB residuals. In the present study, the L/S ratio was increased by 0.11 in the MB-residual compared to the M-residual, which meant that L/S ratio rose by about 50% of the annual L/S ratio (0.2–0.24) without leachate recirculation. Thus leachate recirculation can shorten the time needed to reach the discharge threshold value, e.g., for nitrogen as a previous laboratory study (Höring et al., 1999) have shown that leaching of nitrogen from the landfill in the long term is mainly determined by the L/S ratio. Moreover, in MB residuals the annual (2005) leachate discharge flow was reduced by about 30% probably due to leachate recirculation and increased evaporation in the MB residual compared to the M residual.

In MB landfills, the acidogenic stage is commonly avoided on the evidence of the leachate quality (e.g., pH > 7) found in a laboratory study (Leikam and Stegmann, 1997), in a field-scale lysimeter study (Felske et al., 2003) and in a study of full-scale landfills (Bone et al., 2003) and the gas quality (CH_4/CO_2-ratio) found in a laboratory study (Bockreis and Steinberg, 2005) as well as in a field-scale lysimeter study (Felske et al., 2003). In the present study, with MB residuals a short (2–3 weeks) strong acidogenic phase (pH < 6) was observed after 1 months' leachate flow (after 6 months' landfilling, in May 2005), while transition towards methanogenic phase was observed (pH < 7) in 1–2 months afterwards. On the other hand some VFA (VFA_{cod}/SCOD ratio occasionally 15%) was still present in leachate after 4 months' leachate flow (after 8 months' landfilling), which nevertheless indicated quite a high level of VFA production or a lack of methane-producing bacteria, compared to the VFA_{cod}/SCOD ratio (<3%) 1 month later. Apparently the acidogenic phase was over (BOD/SCOD ratio below 10%) and the methanogenic phase had started – as also supported by the ~50% methane content in the pore gas – after 6 months of leachate flow (10 months' landfilling) in October 2005. However, in practice the acidogenic phase is probably strongly affected by the stability obtained by the MB treatment; thus the acidogenic phase observed in this study may indicate that degree of stabilisation obtained by the MB treatment used was lower than in the MB residual used in previous studies as more than 90% reduce in gas production potential have been reported in previous studies (e.g., Scheelhaase and Bidlingmaier, 1997; Zach et al., 2000) due to biological treatment, while in present study BMP was reduced 80%. On the other hand the use of the VFA_{cod}/SCOD (and BOD/SCOD) ratio and in-situ pH measurements may be a more accurate method of monitoring the degradation stage than the measurement of leachate pH alone. Acidogenic conditions usually increase leachate loads as organic acids are accumulated at higher rate than they are consumed for methane production, which may further result in low and non-optimal pH for methanogens (optimal pH 6.8–7.4 as reviewed by Barlaz et al., 1990). In the M leachate pH remained at about 6 (640 d) and methane concentrations in the pore gas remained at 16–24% (depth of 1.5 m) till the end of 2005. These values together indicate lack of methanogens and/or inhibition of methane production. Furthermore the total VFA concentrations remained high (3500–25,000 mg/l) in the M leachate and a decreasing trend was not observed during the first year landfilling.

The present results show that the methane emissions from the M and MB residuals were low (from 0.05 to 12.4 l/m^2 d in M and from 0.02 to 0.3 l/m^2 d in MB residuals) during the initial phases of landfilling compared to average methane emissions from whole MSW landfills (6–180 l/m^2 d, as reviewed by Kettunen et al., 2006). The

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methane production from MB residuals is likely to be highest in the early phases of landfilling, as shown in another field-scale study (Lorber et al., 2001; Raninger et al., 1999), if the acidogenic phase is avoided due to biological stabilization. Moreover in the present study the small amounts of methane generated in the MB residuals were probably oxidised into carbon dioxide in the cover layer (which was a slightly compacted MB residual). This in turn decreased the methane emissions, as carbon dioxide emissions were at approximately the same level in both residuals, especially during the first 16 months of landfilling. In corresponding landfills without biological stabilisation (M landfills) the acidogenic stage is probably more prolonged due to smaller (<50 mm) particle size compared to acidogenic stage in landfills without pre-treatment of waste and thus methane generation was low in the present M residuals during the initial stage, and carbon dioxide formed the major part of the gas emission. In fact, the M residuals in present study had a high BMP (259 m^3/t TS) compared to the MB residuals (52 m^3/t TS), indicating that after the acidogenic stage, when the methanogenic stage commences, an increase of methane emissions occurs, and may require effective gas recovery and treatment. The SCOD leached from the M residual during the study period (24.2 kg/t TS in 426 days) could contribute to approximately 9.4 m^3 CH_4/t TS (as 1 kg of SCOD can produce 390 l methane at 35 °C, CEC, 1992). Thus part of this SCOD, which will otherwise be lost in the leachate, can be recovered as methane and carbon dioxide, if methanogenic conditions exist. Based on the BMP assay, we estimated (using the CH_4 production by day 21, and on the assumption that methane contributed 60% of the total gas production) that the mean GB21 value of the MB residual used in this study could have been 40–70 Nl/kg TS and not 20 Nl/kg TS as determined for one of the seven batches used in preparing the MB residual. This could indicate e.g. that some of the batches were not well stabilised in the reactor composting phase, as it has been reported (Binner and Zach, 1999) that it is difficult to compensate failures during the first weeks of reactor composting by prolonged composting afterwards. A further explanation could be difficulties in sampling highly heterogeneous materials. Also there were some differences between the BMP and GB21 tests, and we did not measure the GB21 values.

The fate and amounts of leachable compounds in landfills are also affected, aside from the landfill conditions themselves, by the biotic and abiotic factors that take place during the preceding processing/stabilisation stage. Aerobic stabilisation appears to decrease the organic (VS) more significantly than N content of M residual, as indicated by the lower VS/TS ratio of the MB residual (43%) compared to the M residual (61%), while the nitrogen content of the TS were about same in both residuals (0.5% in M and 0.6% in MB) in this study. In previous studies, MB residuals contained approximately same amount (1.1% Cabbai et al., 2005) or less (0.4%, Boni et al., 2006) nitrogen (of TS) than M residuals (1.2%, Cabbai et al., 2005; 1.0%, Woelders and Oonk, 1999) or nitrogen content remained the same (about 1%) during biological stabilization (Heiss-Ziegler and Lechner, 1999). These results (stable nitrogen content% of TS and loss of VS) mean loss of absolute nitrogen content during stabilisation, as part of nitrogen concentrates into a smaller waste mass. On the other hand, it has been reported that absolute nitrogen content can be even more greatly reduced during aerobic stabilisation than in the present and some previous studies; e.g., Boni et al. (2006) reported that 15 days stabilized M residual contained 0.6% nitrogen and after 90 days stabilisation the nitrogen content was reduced to 0.4%. The mechanisms affecting the fate of nitrogen, however, are not fully understood, although a few possible mechanisms, which may affect the fate of nitrogen during MB treatment and landfilling of MB residuals can be suggested (Bone et al., 2003; Cappai et al., 2005). Part of the nitrogen is evaporated as NH_3 (18–1150 g/t treated waste), which can be recovered, e.g., by using scrubbers (Clemens and Cuhls, 2003). Some NH_4–N (as well as organic material) may also be washed out in leachates from composts, thus requiring treatment. The amount of emitted non-methane volatile organic compounds (NMVOC) as gas can rise to 600 g/ton (Soyez and Plickert, 2002), methane from 6 to 8620 g/ton, carbon dioxide from 12 to 185 kg/ton and nitrous oxide from 1.44 to 378 g/ton treated waste (Clemens and Cuhls, 2003) during the aerobic treatment of M residual. On the other hand part of the NH_4–N may also be nitrified during composting (Cappai et al., 2005; Heiss-Ziegler and Lechner, 1999) and denitrified later in landfill conditions (Bone et al., 2003). Moreover composting seems to increase the proportion of humic nitrogen (Cappai et al., 2005; Heiss-Ziegler and Lechner, 1999). For example, the proportion of humic nitrogen rose from about 17% to over 40% during 6 months composting (Ziegler, 1997). It seems that the formation of humic nitrogen might be the major phenomenon, which explains the decreased ammonium nitrogen content and reduced nitrogen load emitted from MB-treated materials (Cappai et al., 2005). In fact in the present study, the leaching of nitrogen and NH_4–N compared to the initial nitrogen content support the notion that nitrogen is strongly compounded – probably as humic nitrogen – into MB residual, as the proportion of leached nitrogen and NH_4–N of total values over 426 d were 1.7% and 1.3%, respectively, from the MB and 14% and 8% from the M-residuals.

5. Conclusions

Landfilled MB residuals cause significantly lower leachate loads of SCOD and NH_4–N, as approximately 40-fold lower SCOD and 5-fold lower NH_4–N loads from the MB compared to M residuals were observed during the 426-day study period. The approximately similar nitrogen content in both residuals and the several-fold lower NH_4–N and nitrogen loads from the MB than M residuals indicated that a major proportion of nitrogen was further stabilised

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during the aerobic phase of stabilisation. Moreover, the leaching of COD, in particular, decreased owing to the shortened acidogenic phase of degradation in the MB residuals. Methane emissions from the MB residuals were low, whereas the methane emissions from the M residuals were up to 20-fold higher than from the MB residuals. However, the methane emissions from the M residuals were low relative to the high BMP in the M residuals, due to the acidogenic phase of degradation. Thus during the methanogenic phase of degradation methane emissions will probably increase strongly and may require effective gas treatment.

Acknowledgements

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