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Landfill Operation and Waste Management Procedures in the Reduction of Methane and Leachate Pollutant Emissions from Municipal Solid Waste Landfills

UNIVERSITY OF ⫿ JYVÄSKYLÄ

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ABSTRACT

The objective of the present research was to find ways of minimising emissions from municipal solid waste (MSW) landfills by means of laboratory experiments. During anaerobic incubation for 237 days, the grey waste components produced between 120 and 320 m^3CH_4 tTS^{-1} and between 0.32 and 3.5 kg NH_4-N tTS^{-1} and the first-order rate constant of degradation ranged from 0.021 and 0.058 d^{-1}. High amounts of COD and NH_4-N were observed in the leachate of grey waste in all the procedures tested during lysimeter experiments lasting 573 days. In the 10-year-old landfilled MSW, a high rate of methanisation was achieved with rainwater addition and leachate recirculation over 538 days, whereas initially pre-wetted grey waste and landfilled MSW were rapidly acidified, thus releasing a high amount of COD into the leachate. In batch assays, the grey waste produced a methane potential amounting to 70-85 % of the total methane potential of the grey waste plus putrescibles. In low moisture conditions, i.e. below 55%, methane production was delayed in the old landfill waste and prevented in the grey waste. In the emission potential study with five waste types, putrescibles produced 410 m^3CH_4 tTS^{-1} and 3.6 kgNH_4-N tTS^{-1}, whereas composted putrescibles produced 41 m^3CH_4 tVS^{-1}, and 2.0 kgNH_4-N tTS^{-1}. The remains of putrescibles probably caused the leaching potential of 2.1 kgNH_4-N tTS^{-1} in the grey waste. Aeration for 51 days in lysimeters reduced the CH_4 potential of putrescibles by more than 68 % and of the lysimeter landfilled grey waste by 50 %, indicating the potential of aeration for CH_4 emission reduction. Nitrogen removal of landfill leachate was studied in the laboratory as well as on-site. Over 90 % nitrification of leachate was obtained with loading rates between 100 and 130 mgNH_4-N l^{-1} d^{-1} at 25°C. Nitrified leachate was denitrified with a loading rate of 3.8 gT.O.N t-$TS^{-1}_{waste}d^{-1}$ without adverse effects on the methanation of waste. In an on-site pilot study a leachate nitrification level of above 90 % was achieved in a crushed brick biofilter with a loading rate of 50 mgNH_4-N/l d even at 5 - 10°C. The effective separation and biological treatment of putrescibles is important in minimising the emission potential of MSW and thus environmental impacts of MSW management. The amount and the rate of release of methane as well as organic matter and nitrogen into leachate can be controlled by active landfill operation. Rapid methanation of MSW by controlling waste moisture and inoculation seems to be the key to achieve low emissions in modern landfills. The controlled access of water and/or leachate can be recommended during the post-closure operation of a landfill. Nitrification in a low-cost biofilter followed by denitrification in a landfill body appears to be feasible method for the removal of nitrogen in landfill leachate in colder climates.

Key words: Bioreactor operation; denitrification; grey waste; hydrolysis; landfill; leachate; municipal solid waste; methane; nitrification; nitrogen solubilisation; source-separation.

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CONTENTS

LIST OF ORIGINAL PUBLICATIONS

The thesis is a summary and discussion based on the following scientific articles, which will be referred to by their Roman numerals I-IV. The studies in papers I-V were carried out by myself and in the papers II, III and V with assistance of Kai Sormunen. The experiments and methodologies in papers II-V were planned by myself in co-operation with Jukka Rintala and Riitta Kettunen, whereas in paper I the experiments and methodologies were planned by myself and Jukka Rintala. The materials for all the papers were compiled, processed and wrote by myself and by the co-authors with a supervision of Jukka Rintala.

I   Jokela, J.P.Y., Vavilin, V.A. & Rintala, J.A. 2002. Hydrolysis rates, methane production and nitrogen solubilisation of grey waste components during anaerobic degradation. Biores. Technol. (submitted).

II   Jokela, J.P.Y., Kettunen, R.H., Sormunen, K.M., & Rintala, J.A. 2002. Methane production from grey waste and landfilled municipal solid waste in a laboratory batch and landfill lysimeter experiment. Environ. Technol. (submitted).

III   Jokela, J.P.Y., Kettunen, R.H., Sormunen, K.M., & Rintala, J.A. 2002. Anaerobic solubilisation of organic matter and nitrogen into leachate from grey waste and landfilled municipal solid waste in a laboratory landfill lysimeter experiment. Environ. Technol. (submitted).

IV   Jokela, J.P.Y., Kettunen, R.H., & Rintala, J.A. 2002. Methane and leachate pollutant emission potential from various fractions of municipal solid waste (MSW): Effects of source separation and aerobic treatment. Waste. Manage. Res. (accepted tentatively and revised).

V   Jokela, J.P.Y., Kettunen, R.H., Sormunen, K.M., & Rintala, J.A. 2002. Biological nitrogen removal from municipal landfill leachate: In situ denitrification and low cost nitrification in biofilters. - Wat. Res. (in press).

VI   Jokela, J.P.Y., & Rintala, J.A. 2002. Anaerobic solubilisation of nitrogen from municipal solid waste (MSW) – a review. Re/Views in Environmental Science and Bio/Technology. (submitted).

ABBREVIATIONS

BMP	biochemical methane potential
BOD_7	biochemical oxygen demand for 7 days
COD	chemical oxygen demand
CPFMSW	composted putrescible fraction of municipal solid waste
DF	downflow filter
DOC	dissolved organic carbon
GHG	greenhouse gas
HRT	hydraulic retention time
MSW	municipal solid waste
L/S	liquid to solids
PFMSW	putrescible fraction of municipal solid waste
SCBP	suspended carrier biofilm process
SCOD	soluble chemical oxygen demand
TKN	total Kjehldal nitrogen
TOC	total organic carbon
TON	total oxidised nitrogen
TS	total solids
UF	upflow filter
VFAs	volatile fatty acids
VS	volatile solids
VSS	volatile suspended solids

1 INTRODUCTION

In many countries the solid waste management is in state of change resulting from the enactment of legislation (e.g. EU Council Directive on the landfill of waste 1999/31/EC) that is passed to promote sustainable development as well as to mitigate greenhouse gas (GHG) emissions by e.g. landfill gas collection and utilisation systems and to minimise the local environmental effects of e.g. landfill leachate. These changes in waste management practice have consequently altered the characteristics of MSW and created new MSW streams, e.g. PFMSW and grey waste. Also, various new waste management and treatment technologies are implemented to limit MSW disposal to landfills and thus limit the emissions from landfills. However, the pollution potential of these waste streams is not well known and thus more information is needed about the effects of the various methods of MSW treatment on the environmental emission potential of the fractions that are landfilled.

Methane emissions from landfills are one of the largest anthropogenic sources of methane and there is global interest in reducing them. The landfills are also sources of water pollutants in the form of landfill leachate that usually contains elevated concentrations of organic matter, ammonium-nitrogen (NH_4-N) (reviewed by e.g. Lema et al. 1988) as well as some organic and inorganic pollutants. The release of organic matter from landfilled municipal solid waste (MSW) is dependant on biological activity as well as on the prevailing physical and chemical conditions, which in turn are influenced by the composition of the landfilled MSW as well as the landfill operating procedures employed (reviewed by e.g. Komilis et al. 1999a).

The main source of nitrogen from landfilled MSW is proteins, which accounts for approx. 0.5 % of dry weight of MSW (e.g. Senior & Balba 1987). The release of soluble nitrogen from municipal solid waste (MSW) into landfill leachate continues over a long period compared to that of soluble carbon compounds (Ehrig 1989). Total Kjeldahl Nitrogen (TKN) content of various MSW from landfill and digestion studies in the literature is presented in Table

1. The TKN content ranges from 1.2 to 4.0 % of TS. Based on the presented TKN concentrations and the amount of the MSW annually landfilled in EU countries (about 136 million tonnes of MSW), around 2 million tonnes of nitrogen is annually disposed to landfills. In Finland the respective amount is around 0.02 million tonnes of nitrogen (about 1.5 million tonnes of MSW).

TABLE 1   Nitrogen contents of the MSW from landfill and digestion studies.

Waste:	TKN (% of TS):	Study:
Landfilled MSW samples	1.2 – 3.8	Landfill (Ham et al. 1993)
Unsorted MSW	3.3	Landfill lysimeter (Pohland et al. 1980)
Unsorted MSW	4.0	Laboratory landfill lysimeter (Leuschner 1989)
Putrescible fraction MSW	3.2	Anaerobic digestion (Cecchi et al.1992)
Putrescible fraction MSW	1.2 - 2.9*	Anaerobic digestion (Gallert & Winter, 1997)
Grey waste fraction MSW	1.2	Laboratory landfill lysimeter (Jokela et al. 2001)

*Calculated by the authors

In-house separation of the PFMSW and its biological treatment is used to reduce the amount of biodegradable waste disposed of in landfills. In addition, recently, various combinations of mechanical and/or biological treatment (reviewed by Komilis et al. 1999b) and/or incineration have increasingly been applied to the treatment of grey waste before its disposal in landfill. At present, both aerobic and anaerobic treatments are used to treat PFMSW and these methods have also increasingly been studied in the stabilisation of grey waste. Moreover, the methanation of organic matter of landfilled waste is potentially controllable by means of various landfill methods, including alteration of the prevailing conditions inside landfills, e.g. by moisture control through leachate recirculation, inoculum and/or pH adjustment (reviewed by e.g. Barlaz et al. 1990, Reinhart & Townsend 1998). An alternative policy in reduction of the landfill emissions is to limit the methanation of waste by preventing the infiltration of water into a landfill by means of an impermeable top cover.

The composition of the MSW is dependant on e.g. waste management legislation and extent of practiced source separation as well as the composition of the materials. Many studies have been done on the composition of unsorted MSW (reviewed by e.g. Barlaz et al. 1990; CEN 1992) and additionally some studies have been done on the characteristics of the individual waste components (e.g. Eleazer et al. 1997). Generally, unsorted MSW has high cellulose content, (10 - 40 % of total solids (TS); Ham et al. 1993). In addition, cellulose and hemicelluloses in waste materials have been shown to be the main source of methane in landfills (Barlaz et al. 1989). The grey waste produced from the source separation may contain high proportion of the cellulose containing waste components, like packaging, cardboard and newsprint, which potentially can yield high amounts of methane if disposed to landfill (Clarkson & Xiao 2000).

Various waste pre-treatments, e.g. source-separation of the putrescible fraction and mechanical-biological treatment of MSW are increasingly carried out before its disposal as landfill. These pre-treatments principally affect various processes in the landfill body, e.g. waste degradation pathways and thus also leachate characteristics. Potentially, the application of source-separation and other pre-treatments reduce the pollution potential of the MSW (reviewed by Komilis et al. 1999b). However, the effect of the source-separation of PFMSW on methane potential production has been poorly studied, although the residual grey waste is still frequently landfilled in many countries. Some studies have reported rather high levels of methane production for the grey waste (e.g. Scherer et al. (1999) reported 0.47 m^3 $kgVS^{-1}$), whereas a lower methane potential has reported for unsorted MSW than for grey waste (e.g. Owens & Chynoweth (1993) reported 0.2 m^3 $kgVS^{-1}$), even if the sorting out of putrescibles should reduce the methane potential of MSW. However, comparison between various studies is difficult owing to the differences between waste management systems and waste characteristics as well as the experimental set-up.

Long-term landfill behaviour and need of after-care is affected by the history of the MSW, including the degradation pathways that have readily taken place and by the landfill technology applied during and after closure of the landfill. Generally, management of a landfill is partly based on the aim of minimising the infiltration of precipitation by constructing an impermeable top cover. On the other hand, higher moisture content (e.g. field capacity level) increases contact between organic compounds and microbes as well as nutrients and thus most likely the rate of waste methanation (reviewed by e.g. Barlaz et al. 1990, Reinhart & Townsend 1998). Some studies (reviewed by e.g. Senior & Balba 1987) have shown an increasing rate of waste methanation with increasing moisture, whereas moisture well above field capacity (approx. 75 %) resulted in sour waste and retarded waste degradation. On the other hand, the rapid stabilisation of MSW through waste methanation would be the preferred method of diminishing the COD load on the leachate. Therefore, the controlled addition of moisture could be an option in increasing the rate of waste methanation and waste stabilisation in landfills.

The rate of production and amount of water pollutants (organic and nitrogen compounds) in leachate during the lifetime of a landfill may be affected by the landfill technology applied e.g. the adjustment of moisture either by waste pre-wetting and/or leachate recirculation (reviewed by Reinhart & Townsend 1998). The degradation pathways may be altered by diminishing the rate of acid production and accelerating methanation by e.g. pH adjustment and the addition of inoculum. Thus, transforming landfill conditions from acidic to methanogenic and thereby diminishing the pollutive load on the leachate is the favoured approach. Pre-wetting of waste prior to landfilling with

the aim of increasing waste degradation rate has been tested, but on the other hand, the elevation of moisture above the field capacity of the waste can increase the acidogenesis of the waste and thus retard methanation (Gurijala & Suflita 1993). Many studies have been done on the effects of leachate recirculation, waste moisture content, and pH adjustment (e.g. Hartz & Ham 1983; Ehrig 1989; Kasali & Senior 1989; Leuschner 1989; Gurijala & Suflita 1993). Active landfill operations have been shown to have positive effects: leachate recirculation has improved leachate characteristics and enhanced waste degradation has been reported to result from e.g. increased moisture, nutrient availability and also from the alleviation of acidity as well as to accelerate waste methanation (reviewed by e.g. Komilis et al. 1999a). Therefore, a combination of leachate recirculation with pH adjustment could minimise the acidity of a landfill and maximise the rate of waste methanation.

The impact of COD pathways (acid production/ methanogenic phase) on nitrogen production in the leachate is a factor that has received little attention (see e.g. review by Burton and Watson-Craik 1998), as the focus of landfill operation studies has mainly been on factors that affect waste methanation. In addition, an elevated nitrogen concentration in leachate from old landfill has also been reported (Trabelsi et al. 2000). Knowledge of the dynamics of nitrogen release during post-closure operation is scarce; however, it is generally known that during anaerobic degradation, carbohydrates, e.g. waste methanation, are preferentially degraded while protein hydrolysis is repressed (Chen et al. 2000). As a result, the nitrogen removal from leachate during post-closure after-care could require a long period of time and thus increase the total operating costs of landfilling to a level much higher than originally expected.

The rate of methanation of MSW in landfills may be limited by the slow growth of cellulolytic microbes (reviewed by e.g. Colberg 1988). These, however, may increase rapidly under aerobic conditions, as demonstrated e.g. in a study on aerobic laboratory MSW lysimeters, which also resulted in high solid destruction (Hanashima 1999). The application of aerobic landfill has been shown to result in lower waste methane generation and leachate pollutant concentration (reviewed by e.g. Read et al. 2001). A 16-week aerobic full-scale composting reduced the respiration rate and methane production potential of grey waste by more than 90 % (Leikam et al. 1997). Lower amounts of ammonium nitrogen and biochemical oxygen demand were found in the resultant leachate produced from the aerobic MSW lysimeter when compared to similar anaerobic landfill lysimeter (Hanashima 1999). Therefore, the aerobic treatment of MSW could be applied as pre-treatment both before anaerobic treatment or landfilling as well as by aeration of existing landfill in order to enhance waste stabilisation.

Currently, waste management produces significant emissions of nitrogen and organic contaminants, as is well known from analyses of landfill leachate

and also rejects from PFMSW treatment plants. The total long-term leachate pollution potential of a landfilled MSW and the effect of mechanical and biological pre-treatment has been studied (reviewed e.g. by Komilis et al. 1999b) and 25-40 gCOD kgTS^{-1} and 1.5-3 gTKN kgTS^{-1} for untreated and 1-3 gCOD kgTS^{-1} and 0.15-0.3 gTKN kgTS^{-1} for pre-treated MSW has been found. Therefore, combining existing source-separation and mechanical pre-treatment with biological (aerobic/anaerobic) processes could be a promising waste management option in diminishing the pollution load of MSW before landfilling.

Normally, the conditions prevalent in a landfill body are anaerobic/anoxic thus enabling methanogenesis and, potentially, denitrification of landfill leachate. Since the aerobic conditions required for nitrification are unlikely to occur in the landfill body, external nitrification is required. Thus, it was concluded that the removal of nitrogen by a process consisting of denitrification in the landfill body preceded by nitrification in a biofilter with low cost filter media, e.g. porous waste material, could be an attractive option. Suspended processes have been applied in full-scale to nitrification and denitrification of landfill leachate (e.g. review by Kettunen 1997). Recently, new methods such as suspended carrier biofilm processes have also been studied for leachate nitrification, even at low temperatures (5 to 10°C; e.g. Welander et al. 1998; Hoilijoki et al. 1999). Previously, the applicability of compost material as a medium for the denitrification and nitrification of landfill leachate has been shown in laboratory scale columns (Onay & Pohland 1998). Anaerobically degrading MSW has also been found to completely denitrify leachate containing nitrate, although the leachate delayed methanogenesis and decreased methane yield (Burton & Watson-Craik 1997), apparently due to suppression of methanogenesis by nitrate (e.g. Chen & Lin 1993; Fang & Zhou 1999; Chidthaisong & Conrad 2000). When planning the use of landfill for denitrification it should be taken into account that denitrifiers may outcompete methanogens (Burton & Watson-Craik 1997).

2 OBJECTIVES

The main objective of this work was to find out the effect of waste pretreatments and characteristics as well as landfill operation procedures on reduction of the methane and nitrogen emissions from municipal solid waste (MSW) landfills.

The source-separated grey waste was characterised by sorting it into main components and sieved into fractions and methane production and nitrogen solubilisation potential of different waste components and fractions were tested. Furthermore, the degradation kinetics of the sorted components was determined and the main sources of methane and soluble nitrogen were traced. (I).

The effects of different waste management and landfill management factors on methane production from the grey waste and from the ten-years-old landfilled MSW were studied with methane production potential assay with PFMSW as a reference and with a batch test to test the effect of moisture on the methane production. Additionally, the effects of the pre-wetting of waste and leachate recirculation with the addition of buffer on the methane production of the two waste types were studied in laboratory landfill lysimeters (II). Also, the effects of these landfill operations on the production of soluble organic matter and nitrogen into leachate was evaluated (III).

Methane and landfill leachate pollution potential of source-separated PFMSW, grey waste, composted PFMSW (CPFMSW) as well as grey waste and the ten-years-old landfilled MSW from laboratory landfill lysimeter experiment were tested and the effects of aerobic treatment on the potentials were evaluated (IV).

A low-cost method for the removal of nitrogen from landfill leachate was developed by studying the nitrification of landfill leachate in laboratory filters with bulking agent of mature compost and crushed brick as the filter material as well as in on-site pilot nitrification study by using a filter of crushed brick. Denitrification of nitrified leachate was studied in a laboratory scale column

containing methane-producing landfilled material and also the effect of denitrification on the methanation of landfilled waste was evaluated (V).

The relevant literature concerning the anaerobic solubilisation of nitrogen from municipal solid waste was reviewed (VI).

3   MATERIALS AND METHODS

3.1 Wastes

Five types of wastes were used, namely source-separated PFMSW, CPFMSW, residual grey waste, landfilled MSW and two samples taken from our previous laboratory lysimeter experiment (Jokela et al. 2001) in which landfilling of grey waste and landfilled MSW was used in the studies (summarised in Table 2). Grey waste was mostly landfilled in Finland around the end of the 1990s when the practice of source-separating PFMSW had been established. Landfilled MSW had been in the landfill for approx. ten years, dated from around the beginning of the 1990s before the era of source-separation.

The grey waste was sampled from Tarastenjärvi waste treatment plant in the Tampere region (Finland). For the characterisation of the grey waste (I), the unprocessed grey waste was delivered in a 1.0 tonne bale wrapped in plastic film. The bale contained approx. 2.5 m^3 of waste and it was in stored at ambient temperature for about one month. Approx. 0.5 m^3 of the waste was shredded to a maximum size of 200 mm which of approx. 0.2 m^3 was sieved with mesh size # 100 mm. This was done to simulate the process carried out in the Tarastenjärvi waste treatment plant. Landfilled MSW was sampled from Ämmässuo landfill in Espoo (Finland) (II, III and IV). The waste samples were excavated from one to ten meters in depth and over an area of about 200 m^2. About 200 l of waste (particle size less than 250 mm) were randomly collected from the excavated material. The samples were shredded with a garden shredder into a maximum particle size of approx. 50 mm and mixed thoroughly. For the batch assay (II), the sample was dried in a constant airflow for one day to remove excess water. No reduction in VS occurred during the drying.

PFMSW and CPFMSW were from Jyväskylä Mustankorkea municipal solid waste treatment plant (Finland) (IV). The PFMSW sample represented kind of PFMSW commonly source-separated in Finland by the end of 1990's,

whereas the CPFMSW sample represented the end product of the main biological method used to treat PFMSW in Finland. For the characterisation study (I), virgin computer printer paper (Canon Office A4, 80 g m^{-2}) was used as a reference. For the BMP study (II) source-separated PFMSW (TS=31.0 % and VS=22.5 %) from the waste treatment plant at Lohja (Finland) was used as a reference, and birch cellulose fiber (TS=94 % and VS=93 %) from the paper mill of Metsä-Serla Oy in Jyväskylä (Finland), was used as a standard. All the samples were milled a maximum particle size of 5 mm in a laboratory with a hammermill (Retsch) before the BMP studies (I and II). The samples from the aeration experiment were milled into maximum particle size of 2 mm for the BMP tests (IV).

TABLE 2   Description and characteristics waste samples.

Waste sample:	Description:	Characteristics:		
		TS (%)	VS (%)	Nitrogen (%-TS)
PFMSW	In-house source-separated PFMSW. Particle size reduced mechanically in laboratory to <50 mm	16	13	0.50
CPFMSW	Composted PFMSW. Static in-vessel composting (forced aeration, RT=7 days) and 6 months windrow composting. Ca. 10 months storing on piles. Sieving out of bulking agent (mesh size # 50 mm). The under-sized for soil amendment and present study.	39	17	0.78
Grey waste	Residual after source-separation of PFMSW (residential areas>6 households), metals, paper, cardboard and glass. Mechanical: magnetic metal removal, shredding (<200 mm), sieving (mesh size # 100 mm), the under-size contained e.g. glass, gravel, ceramics, and some PFMSW, and shredding of the oversize (<50 mm). The sieved undersize and the shredded oversized waste fractions combined in the proportions generated in the plant (68+32 (w+w)).	63	38	1.2
Landfilled MSW	From a closed cell (without methane recovery) containing unsorted MSW from the metropolitan area of Finland from the late 1980s.	42	23	0.63
Lysimeter landfilled grey waste	Grey waste (above) after 573-day laboratory-scale landfill lysimeter study: 148 gCOD kgTS^{-1} 2.7 gTKN kgTS^{-1} and 1.8 gNH$_4$-N kgTS^{-1} removed from the waste as biogas and/or leachate.	48	28	0.68
Lysimeter landfilled MSW	Landfilled MSW (above) after 538-day laboratory-scale landfill lysimeter study: 287 gCOD kgTS^{-1} 0.23 gTKN kgTS^{-1} and 0.11 gNH$_4$-N kgTS^{-1} removed from the waste as biogas and/or leachate.	31	14	0.74

3.2 Sorting test

1.5 m³ of the grey waste was manually sorted into categories (Table 3) that would represent the composition of the grey waste. The category of packaging included all packaging materials that consist of either card alone or of card and layers of other materials such as plastic and/or aluminium. Packaging consisting solely of plastic and other plastic wastes was sorted into their own category. The category of cardboard consisted mainly of brown cardboard boxes and some other cardboard items, such as packaging. The category of textiles included textiles that were made of natural materials, like wool and cotton, as well as of synthetic materials, like polyester. A considerable amount of newsprint as well as diapers were found in the grey waste sample, and these components were thus sorted into their own categories. The component of plastics was not studied for its pollution potential as plastics are considered biologically recalcitrant and thus do not contribute to the produced emissions of organic matter and nitrogen.

3.3 Inocula

Three different digested mesophilic municipal sewage sludge were used as inocula in the studies (I, II and IV). Two different sludges were sampled from Nenäinniemi (Jyväskylä, Finland) with TS of 3.4 % and with VS of 1.8 %) (I) and another one with TS of 2.2 % and with VS of 1.3 % (II). One sludge was sampled from Viinikanlahti (Tampere, Finland) (TS=2.6 %, VS=1.5 %) (IV).

All three laboratory reactors in the nitrification study of the leachate were inoculated with nitrifying activated sludge (4 g VSS (volative suspended solids) l⁻¹) from Viinikanlahti sewage treatment plant in Tampere (Finland) (V).

3.4 Elution test

In the elution test (CEN/TC 1996), the amount of elutable soluble COD and nitrogen was tested with the sorted components and with the fractions of grey waste before and after anaerobic incubation (I) as well as in the five waste samples and in the samples from the aerobic lysimeter experiments (IV). 100 g-TS of sample were first diluted into deionised water in Duran glass vessels to a liquid/solids (L/S)-ratio of 2. The samples were stirred in rotary shaker for 8 h,

which after the liquid samples (approx. 50 ml) were collected. Deionised water was then added to give a L/S-ratio of 8 (overall L/S-ratio of 10). The samples were further stirred in a rotary shaker for 16 h and the liquid samples were collected. All the liquid samples were filtered with glass fibre filters (Schleicher & Schuell). The procedures were performed at 20-22°C. The concentrations after the elution test are expressed as the total cumulative amount of released soluble COD or nitrogen per gTS from L/S=2 and 8 elution.

3.5 Biochemical methane potential (BMP) test

The BMP assays were carried out with the samples from the characterisation study of the grey waste (I), for grey waste and landfilled MSW (II) as well as for the samples from the aeration study (IV). During the BMP assays of the characterisation study, three replicates with the different waste samples at 35°C in 2 l glass vessels for 237 days. The waste samples were added to the vessels to obtain the ratio 1.5 g-VS/gVS$_{inoculum}$ using 0.5 l of the inoculum (I). The BMP assays with grey waste and landfilled MSW were carried out at 35°C in four replicates of 2 l glass vessels for 102 days. PFMSW (0.5 gVS/gVS$_{inoculum}$), grey waste (1.93 gVS/gVS$_{inoculum}$) and cellulose (0.5 gVS/gVS$_{inoculum}$) were added into the vessels with 1 l of the inoculum (II). The different waste materials and those from the aerobic lysimeter experiments were added to obtain 0.5 gVS of waste per gVS of inoculum in the case of PFMSW, and 1.5 gVS of waste per gVS of inoculum in the case of other materials. The milled samples were added to the vessels with 1 l of digested mesophilic municipal sewage sludge (TS=2.6 %, VS=1.5 %) from Viinikanlahti (Finland) as inoculum.

A nutrient solution was added to ensure the availability of micronutrients and trace metals (Lepistö & Rintala 1997), and 5.5 g l^{-1} sodium bicarbonate was added as buffer (I, II and IV). The liquid final volume of 1.6 l in each test vessel was obtained by the addition of deionised water. The inoculum was assayed separately so as to subtract its methane production from those of the samples. The gas phases of the vessels were flushed before the batch assays with N$_2$/CO$_2$ (80 %/20 %) and the vessels were sealed with butyl rubber stoppers. The gas produced was led through Viton tubing to aluminium gas sampling bags (Tecobag PETP/AL/PE-12/12/75 of 10 l, Tesseraux Spezialverpacknungen).

3.6 Batch assay on effects of moisture

The effects of moisture on methane production from the grey and old landfill

waste were studied for 551 and 505 days, respectively (128 gVS and 200 gVS per vessel, respectively) in duplicated 2 l glass vessels at moisture levels of 30, 35, 40, 55 and 75 %-w/w at 22°C±2°C (II). Deionised water was added to obtain the desired moisture level. In both batch studies, the vessels were flushed with N_2/CO_2 (80 %/20 %) and sealed with butyl rubber stoppers. The gas produced was led through Viton tubing to aluminium gas sampling bags (Tecobag PETP/AL/PE-12/12/75, Tesseraux Spezialverpackungen).

3.7   Landfill lysimeter experiment

The lysimeter experiments were run in six parallel lysimeters made of 150 mm PVC pipes (height 600 mm) sealed with PVC flanges and with an empty volume of 8.8 l (II and III) at 22°C±2°C. The leachate produced was discharged from the bottom of the lysimeters through Tygon tubes to 1 l Duran glass vessels sealed with rubber stoppers and placed in a refrigerator (4°C). The biogas produced was collected through a gas tube (Viton) in aluminium gas sampling bags after passing a water lock. Three lysimeters were filled with grey waste (4.6 kg in each; referred to as L1-L3) and three with old waste (7.5 kg in each; referred to as L4-L6) and they were run for 573 and 538 days, respectively (Table 1). The difference in the amounts of waste was due to the compaction characteristics of the respective wastes. The headspaces of the lysimeters were flushed with N_2/CO_2 (80 %/20 %). At the start of the experiment, one lysimeter (L3 and L6) of each of the waste types was saturated with deionised water for 3 days and then drained. 3.97 l was leached immediately out of grey waste lysimeter (L3), whereas excess water of approx. 2.0 l was leached out of the landfilled MSW over 6 weeks. The calculated moisture concentrations after draining were 55 % and 66 %, respectively, and thus the field capacity of the grey waste was 48 % and that of the MSW 60 %. Deionised water with a volume (220 ml week^{-1}) equal to the mean weekly local precipitation (per lysimeter surface) was added weekly to the lysimeters. Leachate production started immediately in the pre-wetted lysimeters (L3 and L6), whereas in grey waste lysimeters it started on days 44 (L1) and 58 (L2) and on day 23 in the other MSW lysimeters (L4 and L5). Leachate recirculation (220 ml week^{-1}, added simultaneously with deionised water) was started on experimental day 58 in L2, L3, and on day 23 in L5 and L6, when the materials in L2 and L4 (and L1 and L3) were at field capacity. Before recirculation, the pH of the leachate was adjusted to 7.0 (L2, L4) or 8.0 (L3, L6) with 1 M NaOH. By the end of the lysimeter experiments, the liquid/solids (L/S) ratios were 4.5 and 8.7 in the control lysimeters and L2, L5 and L6, respectively, whereas in the pre-wetted grey waste lysimeter (L3) the ratio was 11.7. On the basis of the known levels of

water infiltration at the local prevailing conditions on landfills (waste density 500 kgTS m^{-3}, average annual precipitation 650 mm), the L/S ratios of 1 and 5 would be the equivalent of time periods of 7.5 and 40 years, respectively.

3.8 Aerobic lysimeter experiment

The aerobic treatment was studied in five parallel lysimeters made of 600 mm PVC-pipes (diameter 150 mm and total volume 9.4 l) and sealed with PVC-flanges (IV) at 25°C. The lysimeters were filled to an equal height of 450 mm with the PFMSW (4.5 kg), CPFMSW (5.2 kg), grey waste (2.5 kg), lysimeter landfilled grey waste (3.2 kg) and lysimeter landfilled MSW (4.0 kg) samples. All lysimeters were aerated from the bottom through an air diffuser, which was covered with a 50 mm layer of grit (grain size from # 5 to 15 mm). The gas tubing material used was PVC. The tops of the lysimeters were sealed with PVC flanges to prevent desiccation of the waste. The gas was passed through an outlet of the cover into a water lock to maintain equal counter pressure in all lysimeters and to prevent channel flow.

At the beginning of the experiments, the airflow rate for the PFMSW was set higher (approx. 0.2 m^3 d^{-1} kg^{-1}) than for the other materials (approx. 0.08 m^3 d^{-1} kg^{-1}) owing to its presumably higher aerobic degradation rate. These flow rates were based on previous findings indicating that higher flow rates can cause desiccation of waste (e.g. review by Stessel & Murphy 1992). Later on during the experiment, the airflow rate in all the lysimeters was adjusted to ensure the O$_2$ content of the outflow gas (measured every weekday) was above 15 % and the CO$_2$ content above the detection limit (i.e. 0.1 %). Consequently, the airflow rates through the lysimeters were reduced by the end of the experiment to 0.03 and 0.015 m^3 d^{-1} kg^{-1}, respectively. The waste materials were sampled at start-up (day 0) and on days 23 and 51 (the end of the experiment) directly from the top of lysimeter.

3.9 Landfill leachate nitrification and denitrification experiments

3.9.1 Landfill leachate

Leachate was sampled for laboratory experiments from the main drain at Mustankorkea municipal landfill in Jyväskylä (Finland), which has been in operation since 1963 (V). The total volume of landfill in 1999 was approx. 5 million m^3 and the annual production of leachate was ca. 114000 m^3. In addition

to MSW, some industrial waste (e.g. fiberwastll from a papermill, fly ash and waste grit) has been disposed of in the landfill. Putrescible household waste has been source sorted and composted separately since 1996. The leachate was delivered 1-2 times per month to the laboratory and stored at 4°C. The feed for the experiments was prepared by clarifying the leachate in a container. Due to a deficiency of phosphorus in the landfill leachate, 5 to 20 mg phosphorus per litre (1 M H_3PO_4) was added to the feed. The ammonium nitrogen (NH_4-N) of the feed was typically 60 to 170 mgN I^{-1}, biochemical oxygen demand (BOD_7) was 18 mgO$_2$ I^{-1} and the chemical oxygen demand (COD) was 230 to 510 mgO$_2$ I^{-1}. Between experimental days 135 and 156 of the runs, the leachate contained some exceptional streams from the landfill and, consequently, the feed had a higher COD (1300 mgO$_2$ I^{-1}) and BOD_7 (510 mgO$_2$ I^{-1}), whereas ammonium remained between 60 and 90 mgN I^{-1}. During the nitrification experiment, the pH tended to rise above 8.5 resulting from loss of carbonate, which may cause loss of ammonia by volatilisation. The feed pH was decreased by the addition of 1 M hydrochloric acid (HCl) from approx. 7.0 to 6.5 to maintain the pH of the effluent below 8.5. Identical feed was used in all the nitrifying laboratory reactors. During the runs, the ammonia load was mainly controlled by adjusting the HRT, but it was also affected by the changes in the feed ammonia concentration.

The leachate used in the pilot study was pumped from the main leachate ditch at Kiertokapula municipal landfill in Hyvinkää (Finland). The landfill has been in operation since 1974 and approx. 1.3 million m^3 of MSW has been disposed of in the landfill. Also at this site, source separated putrescible household waste has been composted separately since 1996. The drainage of the compost plant is discharged into the landfill and was thus included in the leachate used in the experiment. The availability of soluble phosphorus was ensured (COD:P ratio of 100:1) by the continuous addition of 10 to 30 mg phosphorus (1 M H_3PO_4) per litre to the leachate in a mixing tank. The NH_4-N of the leachate varied between 160 and 270 mgN I^{-1} (NH_4-N/TKN-ratio was between 0.78 and 0.99) and the COD between 1300 and 1600 mgO$_2$ I^{-1} and the BOD_7 was 5 mgO$_2$ I^{-1}. The pH varied between 6.8 and 7.5.

3.9.2 Laboratory-scale experiments

3.9.2.1 Nitrification

Three types of reactors were used in the laboratory-scale experiments for nitrification of landfill leachate: an UF with crushed brick as the filter medium, a DF with a bulking agent (wood chips) of mature compost of source-separated putrescible household waste as the filter medium and a SCBP with a commercial carrier (V). Nitrified leachate from the SCBP was subsequently fed

into a denitrification landfill column filled with two separate horizons of old landfilled waste of different ages. All the laboratory-scale experiments were carried out at ambient temperature (25°C). After the laboratory experiment, an appropriate method was selected for the nitrification pilot study, which was carried out on a landfill site. The nitrification filters were operated in all experiments with a loading rate between 25 and 130 mgNH$_4$-N g l^{-1} d^{-1}, which was considered sufficiently high for the actual cases. No maximum loading rate of the different processes were studied

The leachate was continuously pumped from the feed container (4°C) with a peristaltic pump (Cole Palmer, Masterflex 7553-87) into the nitrification reactors. The UF and DF were made of 600 mm PVC pipes (total volume 9.4 l) sealed with PVC flanges. In the UF, the leachate was fed through a 50 mm layer of grit (grain size from # 5 to 15 mm) to the bottom of the filter. The void volume of the UF was approx. 2.4 l. In the DF filter, the leachate was distributed to the top of the reactor through a perforated plate. The DF medium was first washed with tap water and sieved into particles of 10 to 70 mm. At the beginning of the experiment, the nitrifying compost column was rinsed with 8 l of tap water to reduce further the amount of soluble material. The SCBP was made of a 400 mm PVC pipe (total volume of reactor 7.1 l). The bottom of the SCBP was sealed with a PVC flange and the top covered with aluminium foil to minimise evaporation. 40 % of the reactor volume was filled with polyethylene carrier medium (Kaldnes Miljöteknologi AS, Norway, bulk density = 170 kg m^{-3}, diameter = 9.1 mm and length = 7.2 mm) with an effective surface of 500 m^2 m^{-3}. PVC was used as the tubing material in all the nitrification reactors. All three reactors were inoculated with nitrifying activated sludge (4 g VSS (volative suspended solids) l^{-1}) from Viinikanlahti sewage treatment plant in Tampere (Finland) and were aerated with submerged diffusers (Rena Air 200). On account of the mixing requirements, dissolved oxygen in the SCBP reactor was approx. 8 mg-O$_2$/L.

3.9.2.2 Denitrification

The denitrification column was made of 600 mm PVC pipe (column volume 8.8 L) sealed with PVC flanges (V). 220 ml of nitrified leachate from the SCBP was fed to the top of the column with a syringe once a week. The TON loading rate was calculated from the weekly-added nitrified leachate. An identical column was operated without added leachate and was used as a reference. Both columns were filled with two layers (250 mm) of different landfill waste. The upper layer consisted of landfilled MSW with a maximum particle size of 50 mm. The characteristics of the waste (753 gTS and 345 gVS in column) are reported elsewhere (Jokela et al. 1999). The bottom layer consisted of ca. 45-years-old MSW (657 gTS and 104 gVS in column) from Mattilanniemi landfill in

Jyväskylä (Finland), which has been closed for approx. 40 years. The 50 mm layer of grit on the bottom of the columns prevented washout of particles. The leachate from the denitrification column was led from the bottom of the column through Tygon tubing to 1 L Duran glass vessels (at 4°C), which were sealed with rubber stoppers. The gas produced was led through Viton tubing to the gas sampling bags (Tecobag PETP/AL/PE-12/12/75 of 10 I, Tesseraux Spezialverpacknungen) after passing a water lock.

3.9.3 Pilot

3.9.3.1 Nitrification

In the pilot study the upflow nitrification filter (pilot UF) was constructed from a 2000 mm long HDPE pipe (diameter 920 mm and a total volume 1.0 m^3) sealed with a HDPE flange (V). Crushed brick was used as the filter medium. The medium was sieved into particles of 16-32 mm. The influent was fed through a perforated plate and the effluent was discharged from an overflow outlet. The leachate was semi-continuously pumped from the main leachate ditch with a submersible pump into a 2 m^3 storage tank, which from it was led to a 100 I mixing tank in which phosphorus was added. The leachate was continuously pumped from the mixing tank with Watson-Marlow (504 S/313D) peristaltic pump via a PVC tube into the reactor. A diaphragm pump (KNF) was used for aeration. At the start-up of the experiment, the reactor was inoculated with nitrifying activated sludge (2 gVSS l^{-1}), which was supplied from Hyvinkää sewage treatment plant (Finland). During the experiment, dissolved oxygen in the filter (from depth of 200 mm) was between 4.2 and 9.0 mgO_2 l^{-1}. The filter was operated for the first five days on batch mode to ensure the establishment of microbial biofilm on the filter medium.

3.10 Analyses

Biogas volume was measured using a displacement method. The methane concentration was analysed with a Perkin Elmer Autosystem XL gas chromatograph, equipped with a flame-ionisation detector and fitted with a PE Alumina column (30 m x 0.53 mm). The CO_2 and O_2 concentration were analysed with an IR-analyser (Geotechnical Instruments) (I, II, IV and V). Leachates were sampled weekly from effluent of the lysimeters, and volume, pH, soluble COD, and ammonium-nitrogen (NH_4-N) were analysed immediately. Other samples were stored in PVC bottles at -20°C (III).

pH was measured in the laboratory with a Metrohm model 744 pH meter,

whereas during the pilot experiments, pH was measured with a Hanna model HI 9025 pH meter (V). The dissolved oxygen was measured with a Hanna Instruments HI-9145 oxygen meter. Volatile fatty acids (VFAs from C_2 to C_6) were analysed with a Perkin Elmer Autosystem XL gas chromatograph, equipped with a flame-ionisation detector and fitted with a PE FFAP column (30 m x 0.32 mm x 25 μm). Ammonium nitrogen (NH_4-N) and T.O.N were determined by using a Kjeltec system 1002 distilling unit (Perstorp Analytical/Tecator AB) in accordance with *Standard Methods* (APHA 1998). Total Kjeldahl nitrogen (TKN) analyses were done by following the Tecator application procedure (Perstorp Analytical/Tecator AB, 1995) with the same Kjeltec system after digestion with a 2006 digestor. Nitrate and nitrite nitrogen as well as phosphate phosphorous were measured from filtered samples (0.2 μm filters Schleicher & Schüll) with Dionex DX 500 ion chromatograph. COD (dichromate method) and BOD_7 were analysed according to the Finnish Standards Association (SFS 5504 1988; SFS 5508 1991). Total organic carbon (TOC) and dissolved organic carbon (DOC) were analysed with a Schimadzu 5000 according to the CEN 1484 method (CEN 1997).

TS and VS were analysed according to Standard Methods (APHA 1998). Cellulose concentration was measured by a method based on sulphuric-acid digestion (Safarik & Santruckova 1992), after which the concentration of glucose in the liquid phase was measured spectrophotometrically (Perkin Elmer Lambda 5 UV/VIS Spectrophotometer) (Dubois et al. 1956). In this method, starch is accounted as glucose. The amount of starch was expected to be low as MSW usually contains less than 1% starch. COD (dichromate) was analysed according to *Finnish Standards* (SFS 5504 1988). The amount of immobilised biomass in the SCBP was determined by mechanically scraping biomass from the surface of twelve carriers and determining TS and VS of the scraped biomass.

3.11 Calculations and data analyses

The cumulative methane yield of each waste material was calculated by dividing its cumulative methane production by the amount of waste added (TS and VS) to each vessel (I). The rate of degradation of the materials was assumed to follow a first order kinetic rate of degradation, and thus the following formula was used to describe the methane production rate of each category of material:

$$Y = Y_\mu * (1 - \exp[-k * t]),$$

where Y represents the cumulative methane yield at time t, Y_μ represents the ultimate methane yield of the sample and k is the first order rate constant. The parameters Y_μ and k were estimated by using non-linear regression fit to the yield data of a triplicate set with SPSS for Windows 10.1.3 software (I).

The L/S ratio was calculated by dividing the amount of water or leachate applied by the initial TS content of the waste in the lysimeter. The possible period needed to achieve these concentrations in the leachate in the actual landfills can be estimated in relation to the L/S ratio, providing that the local landfill conditions in terms of e.g. water infiltration and waste depth and density are known (III).

The maximum methane production rate per gram of added inoculum as VS was estimated by linear regression analysis of the steepest slope in the cumulative methane production curve (IV). In the lysimeter studies, total released COD was the sum of the methane COD (350 mlCH$_4$ $\cong$ 1 gCOD at NTP) and the COD in the discharged leachate (II).

The nitrification efficiency was calculated by dividing the effluent NO$_3$-N by the influent NH$_4$-N and multiplying it with 100 % (V).

4 RESULTS AND DISCUSSION

4.1 Composition of grey waste

The composition and the characteristics of the waste fractions in the source-separated grey waste as received at the waste management plant are presented in Table 3 (I). It showed that, along with the remnants of putrescibles, a high amount of other biodegradable waste components were present in the grey waste, including packaging cartons (4.4 %) and cardboard (10 %), newsprint (11 %), textiles (12 %) and diapers (3.6 %) comprising 41 % of the grey waste. Previously, in a similar sorting test with grey waste the proportion of the same components was 40 %, made up of 16 % of various packaging materials (multilayer cartons, cardboard and paper), 5 % newsprint, magazines and office paper, 8 % textiles as well as 9 % diapers (Huotari 1994). In the same study the plastics contributed 11 % of the grey waste, and putrescibles 26 %. Putrescibles were omitted in the present study, but the miscellaneous unsorted fraction probably contained residuals of putrescibles, as in packaging, which is often contaminated with putrescibles. For unsorted MSW the proportion of non-putrescible biodegradable waste has been between 20 and 42 % (reviewed by CEC 1992; reviewed by Barlaz et al. 1990).

TABLE 3   The composition of the residual grey waste.

Grey waste component:	(%-w/w)	TS (%):	VS (%):	VS/TS:	Soluble fraction (COD % of TS)	TKN (% of TS):	Elutable NH$_4$-N (% of TKN)
Packaging	4.4	84	72	0.85	0.02	0.03	23
Cardboard	10	87	67	0.77	0.02	0.04	100
Textiles	12	91	84	0.92	0.01	0.06	19
Newsprints	11	86	59	0.68	0.01	0.06	0
Diapers	3.6	38	29	0.78	0.09	1.5	7.2
Plastics	15	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Oversized	68*	85	70	0.82	0.01	0.25	4
Undersized	32*	78	54	0.69	0.02	0.32	16
Grey waste	100	84	58	0.69	0.02	0.32	29
Sorted fractions	56	-	-	-	-	-	-
Miscellaneous unsorted	44	-	-	-	-	-	-
Office paper	-	97	72	0.74	0.02	0.02	n.d.

* Data supplied by the waste management company
n.a.: Not analysed

4.2 Biochemical methane potential of source separated MSW fractions and hydrolysis rates of grey waste components

The specific methane yields (per TS and VS) together with the estimates of the ultimate methane yield (Y_μ) and the first order rate constant of each grey waste material after 66 days (I) and from those prepared in a previous study (IV) after 110 days with the putrescibles are presented in Table 4. The methane production began within 3 days in all samples and most of the residual grey waste components (packaging, cardboard, textiles and diapers) seem to yield high amounts of methane within days 43 and 66 in the presence of a proper methanogenic inoculum. Previously, in a similar BMP study, methane yields of between 318 and 343 m^3 tVS^{-1} were obtained for packaging materials, including milk cartons, wax paper and food board (Owens & Chynoweth 1993). The larger maximum particle or the protective film on cartons and cardboard in the waste in the present study may have impeded the hydrolysis of solids by decreasing the access of hydrolytic enzymes to the cellulose-containing waste components. The high concentration of inorganic clay filler can be seen in the somewhat lower VS content in the newsprint and in the office paper if compared to the content in the packaging. Therefore, the characteristics of the waste component, e.g. the method of paper and carton production used, effects the rate and extent of methanation and destruction of solids. Nevertheless, the high cellulose-containing components of the grey waste will eventually degrade and yield high amount of methane. Methanation may thus be applied as a biological method of treatment before landfill disposal of the grey waste.

The methane production potential of the grey waste as well as of the PFMSW and cellulose were studied in the BMP assays (Table 4) (II). Most of the methane was produced from the materials within 11 days (Fig. 1a). The methane yield from the grey waste was about half of that of the PFMSW when calculated per wet weight of waste, and approx. one quarter when calculated per TS or VS. Thus, grey waste may still produce significant amounts of methane despite in-house separation of PFMSW, if an appropriate inoculum is available and/or if moisture and other conditions (e.g. neutral pH) in the landfill are favourable. It can be estimated that grey waste would have a methane potential amounting to 70-85 % of the total methane potential of the grey waste plus source-separated PFMSW. Therefore, the residual grey waste has to be further treated mechanically and biologically to reduce the gas formation e.g. below 20 ml g TS^{-1} within 21 days (Anon 1997).

The high coefficients of determination (above 0.88 with the methane yields of all the individual vessels (values not shown) indicated that the first order kinetic model described rather well the degradation of each grey waste fraction. In a previous study (Owens & Chynoweth 1993), slightly higher constant was obtained for the waste components, e.g. newsprint between 0.069 and 0.084 d^{-1} and corrugated unbleached cardboard 0.058 d^{-1}, whereas the constants for packaging (between 0.099 and 0.119 d^{-1}) and especially for office paper (0.136 d^{-1}) were clearly higher than the present ones. This might result from differences between the studies in terms of the types and amounts of plastic films (packaging) used or amount of clay filler (office paper) in the waste components, which in present study might have decreased the transfer of mass from the waste particles. The range of the constant for putrescible components has been reported to be between 0.10 and 0.35 d^{-1} (Veeken & Hamelers 1999).

A high amount of nitrogen (Table 5) was solubilised in the form of NH_4-N from the undersized fraction of the grey waste during methanation resulting probably from the remnants of protein-containing putrescibles. 1.0 gNH_4-N kgTS^{-1} was eluted from the grey waste after the 237-day anaerobic incubation experiment. The great differences in NH_4-N yield between the oversized and undersized fraction suggest that the presently applied separation of fractions by sieving is an effective method of reducing the nitrogen load from the waste management if the undersized fraction is properly treated and end-disposed.

TABLE 4   The specific methane yields of the grey waste components during the 237 days of the biochemical methane potential test (BMP) and from the grey waste, source-separated PFMSW and cellulose during the 102 days of the biochemical BMP test at 35°C with added methanogenic inoculum and the estimated values of the ultimate (Y_μ) and the first-order kinetic constant (k) (standard deviation in parenthesis).

Grey waste component:	Specific methane yield			Y_μ (m³ tTS$^{-1}_{added}$)	k (d^{-1})
	(m³ t $_{waste}$$^{-1}_{added}$)	(m³ tTS$^{-1}_{added}$)	(m³ tVS$^{-1}_{added}$)		
Packaging	118(81)	140(96)	165(112)	150 (100)	0.058 (0.01)
Cardboard	146(25)	168(29)	217(38)	183 (32)	0.046 (0.009)
Textiles	192(99)	210(109)	228(118)	238 (121)	0.021 (0.001)
Newsprint	34(6.1)	40(7.1)	58(10)	43.6 (7.0)	0.056 (0.008)
Diapers	60(5.1)	158(13)	204(18)	177 (14)	0.025 (0.003)
Oversized	127(26)	150(31)	182(38)	166 (35)	0.031 (0.003)
Undersized	118(5.4)	151(6.9)	219(10)	170 (7.3)	0.026 (0.002)
Grey waste	85(2.4)	101(2.8)	147(4.1)	112 (3.8)	0.031 (0.001)
Office paper	243(18)	252(18)	340(24)	277 (21)	0.036 (0.006)
*Putrescibles	67(6.9)	410(42)	527(54)	426 (42)	0.107 (0.04)
Grey waste	64 (13)	100 (20)	140 (29)	n.a.	n.a.
PFMSW	130 (0.66)	440 (2.2)	520 (2.6)	n.a.	n.a.
Cellulose	570 (1.1)	610 (1.2)	610 (1.2)	n.a.	n.a.

*From a similar batch assay (IV)

TABLE 5   Reduction of solids (TS and VS), VS/TS and specific NH_4-N solubilisation from the grey waste and its components (inoculum subtracted) as well as NH_4-N/TKN ratio of samples (inoculum included) after the 237 days of the biochemical methane potential test (BMP) at 35°C with added methanogenic inoculum (standard deviation in parenthesis).

Grey waste component:	$TS_{red.}$ (% of waste$_{added}$):	$VS_{red.}$ (% of waste$_{added}$):	VS/TS (final of waste):	NH_4-N/TKN (of waste+inoculum):	Specific NH_4-N solubilisation (gN kgTS$^{-1}_{added}$)
Packaging	52(7.6)	52(4.7)	0.46(0.03)	0.53	<0.01
Cardboard	50(4.1)	54(12.8)	0.43(0.09)	0.52	<0.01
Textiles	70(3.1)	79(2.9)	0.37(0.2)	0.51	<0.01
Newsprint	29(1.3)	27(1.0)	0.47(0.01)	0.49	<0.01
Diapers	40(1.2)	43(2.3)	0.46(0.02)	0.69	9.8 (0.44)
Oversized	49(4.1)	51(3.5)	0.45(0.01)	0.9	0.32 (0.05)
Undersized	31(1.1)	35(0.4)	0.45(0.02)	0.59	3.5 (0.25)
Grey waste	33(1.1)	35(2.0)	0.45(0.01)	0.58	1.0 (0.003)
Office paper	73(0.6)	88(2.7)	0.31(0.01)	0.56	<0.01

4.3 Effect of moisture on methane production

The effect of waste moisture on methane production in the grey and old landfill waste was studied in the batch assays without the addition of inoculum (Table 6 and Fig. 1b) (II). The grey waste produced methane only at the highest moisture concentration (75%) whereas the old landfilled waste produced methane promptly in all moisture concentrations and had the highest methane yields (54 mlCH$_4$ gTS^{-1}) at 55 % and 75% moisture while the grey waste at 75% moisture had the highest specific methane yield (77 mlCH$_4$ gTS^{-1}). At the lowest moisture levels (30 and 35 %), the old landfilled waste produced almost an identical amount and rate of methane, whereas at 40 % moisture both the rate and the amount of methane production were somewhat higher. Thus, a low waste moisture level significantly suppresses (old landfilled waste) or prevents (grey waste) the methanation of waste. These results together with the results of the studies by Gurijala & Suflita (1993) suggest that moisture content close to the field capacity of waste (approx. 50 %), may be optimal for unsorted landfilled waste, resulting in faster methanation and three- to fourfold higher methane yields during the study periods than at 20 to 30 % moisture content.

In the grey waste the low moisture content suppressed the initiation of methanation more and the lower methane yield may have resulted from the accumulation of inhibitory intermediate compounds leading to acidic conditions due to the shortage of methanogens. In an old landfill without moisture control, the moisture content of waste may vary from 15 to 50 % in different parts and depths of the landfill (e.g. Reinhart & Townsend, 1998) and it would probably be at the lower end of those values in post-closure landfill with an impermeable top cover if moisture addition, e.g. leachate recirculation, is not applied. On the other hand, in our study the slow continuation of methane production even in low moisture conditions in old landfilled waste suggests that methane will be produced, although at very much decreased rates, as reported by Hartz & Ham (1983). Thus, in a landfill, low waste moisture concentration due to e.g. the prevention of water infiltration through the use of an impermeable top cover would apparently not completely prevent methane production, but rather lead to methane production over a longer period.

TABLE 6   Effect of moisture on methane yield from the grey and old landfill waste in the batch assay without added inoculum at 25°C. Standard deviation in parenthesis.

Substrate/initial moisture	Specific methane yield		
	mlCH$_4$ g$_{feed}$$^{-1}$	mlCH$_4$ gTS^{-1}	mlCH$_4$ gVS^{-1}
Grey waste			
75 %	48 (0.9)	77 (1.4)	130 (2.3)
Old landfill waste			
30 %	9.7 (0.1)	14 (0.1)	21 (0.2)
35 %	13 (0.4)	21 (0.7)	33 (1.1)
40 %	18 (0.4)	28 (0.6)	44 (0.9)
55 %	35 (0.7)	54 (1.1)	84 (1.8)
75 %	34 (0.7)	54 (1.2)	83 (1.8)

Run time (days)
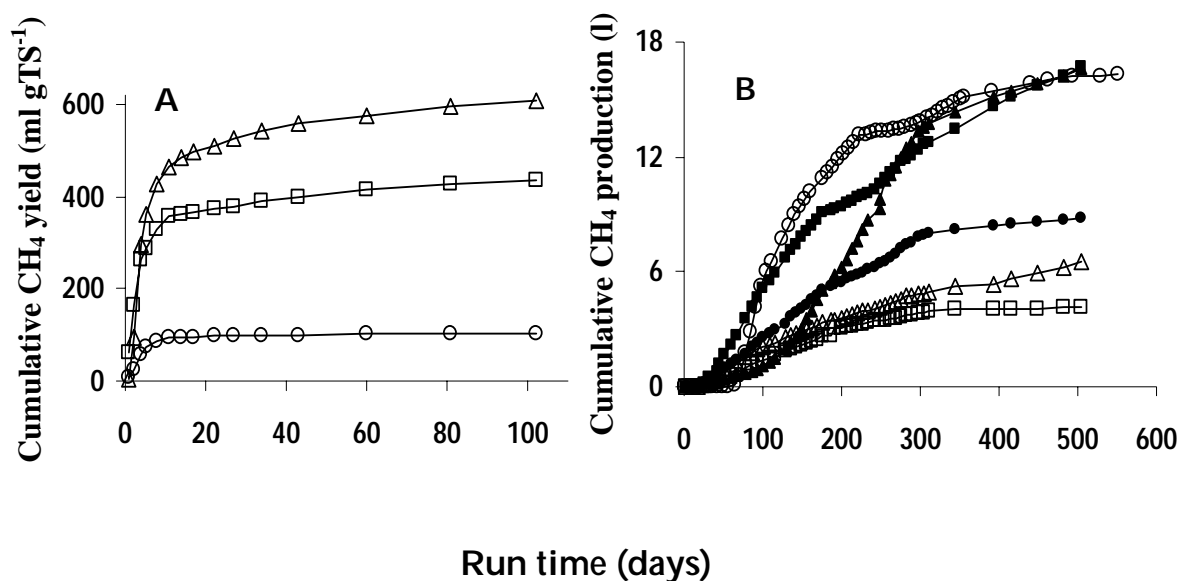
FIGURE 1   Methane yields from the grey waste (’) PFMSW (œ) and cellulose ( ) in the BMP test at 35°C with added methanogenic inoculum (a) and effect of waste moisture on cumulative methane production from the grey (’) and the old landfill waste (30 œ, 35 , 40 ‘ , 55 › , 75% d moisture) in the batch assay at 25°C without added inoculum (b).

4.4 Landfill bioreactor operations

4.4.1 Effect of waste pre-wetting and leachate recirculation with pH adjustment on methane production and on leachate characteristics

The effects of initial waste pre-wetting and leachate recirculation with pH adjustment on the methane content of biogas and methane production were studied studied in the lysimeter experiment and also in the reference lysimeters without recirculation (II) (Fig. 2). Total COD release via gas and leachate and on CH_4-COD versus total COD release and on specific methane yields (Table 7) were determined on the basis of the experiment. In the grey waste the methane content of the biogas increased towards the end of the experiment in all lysimeters, but the methane production remained low (Fig. 2). The pre-wetted material (L3) released clearly more COD via leachate than the other recirculated lysimeter (L2). This was apparently due to retarded development of methanogenic activity in the prevailing acidic conditions, as indicated by the low pH and high VFA in the leachate (III). In the landfill-scale, a lower rate of infiltration of moisture, and consequently a lower waste moisture content than that investigated in the present study as well as the greater heterogeneity of the landfill waste would be more likely to limit contact between the waste and micro organisms. In practice, it would take a long time to dilute the acidic conditions. Therefore, a landfill should be operated more in the manner of bioreactor (Reinhart & Townsend 1998) and more drastic measures, like the addition of inoculum in the form of e.g. digested sludge into the waste (reviewed by e.g. Barlaz et al. 1990), should be applied to initiate waste methanation and thus prevent a long lasting production of high strength leachate.

The prompt methanation of MSW without pre-wetting was possible due to the level of methanogenic activity established before the experiment, but the acidity in the pre-wetted one effectively retarded methanation. Previously, on the basis of a laboratory leach-bed study with unsorted MSW (Lai et al. 2001), after the onset of sustained methanogenesis a high SCOD was hydrolysed and released as methane and thus waste methanation seems to promote hydrolysis. The delayed methane production and high COD of the leachate from the initially pre-wetted old waste was apparently caused by the rapid acidification of the waste. Therefore, pre-wetting may imbalance the consumption and production of intermediate products and thus retards the waste methanation. Previously, 7.5-fold COD release was obtained from an initially pre-wetted waste lysimeter with high cumulative methane production (Doedens & Cord-Landwehr 1989). In practice, the range of moisture concentrations applied (pre-wetting vs. no pre-wetting) would most likely coexist in a landfill and thus the

inhibiting intermediates would probably be consumed in different layers of the landfill, especially if leachate recirculation is applied.
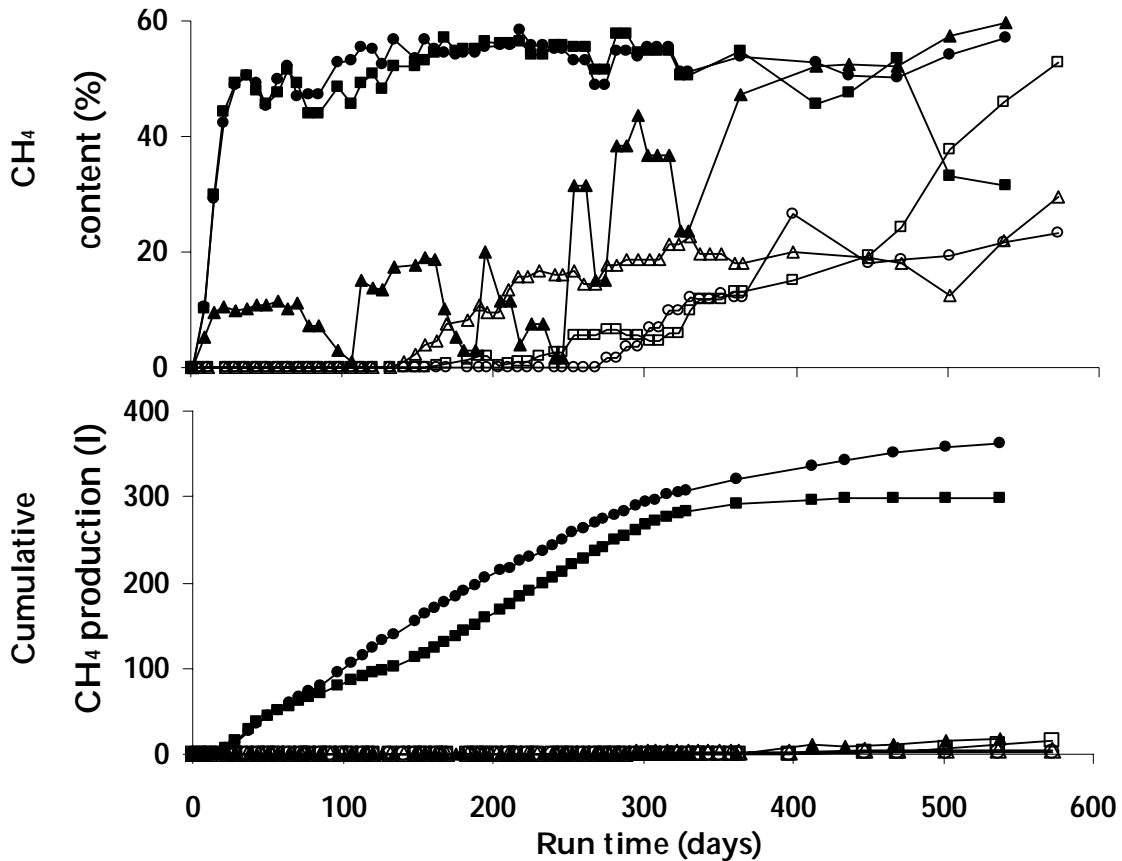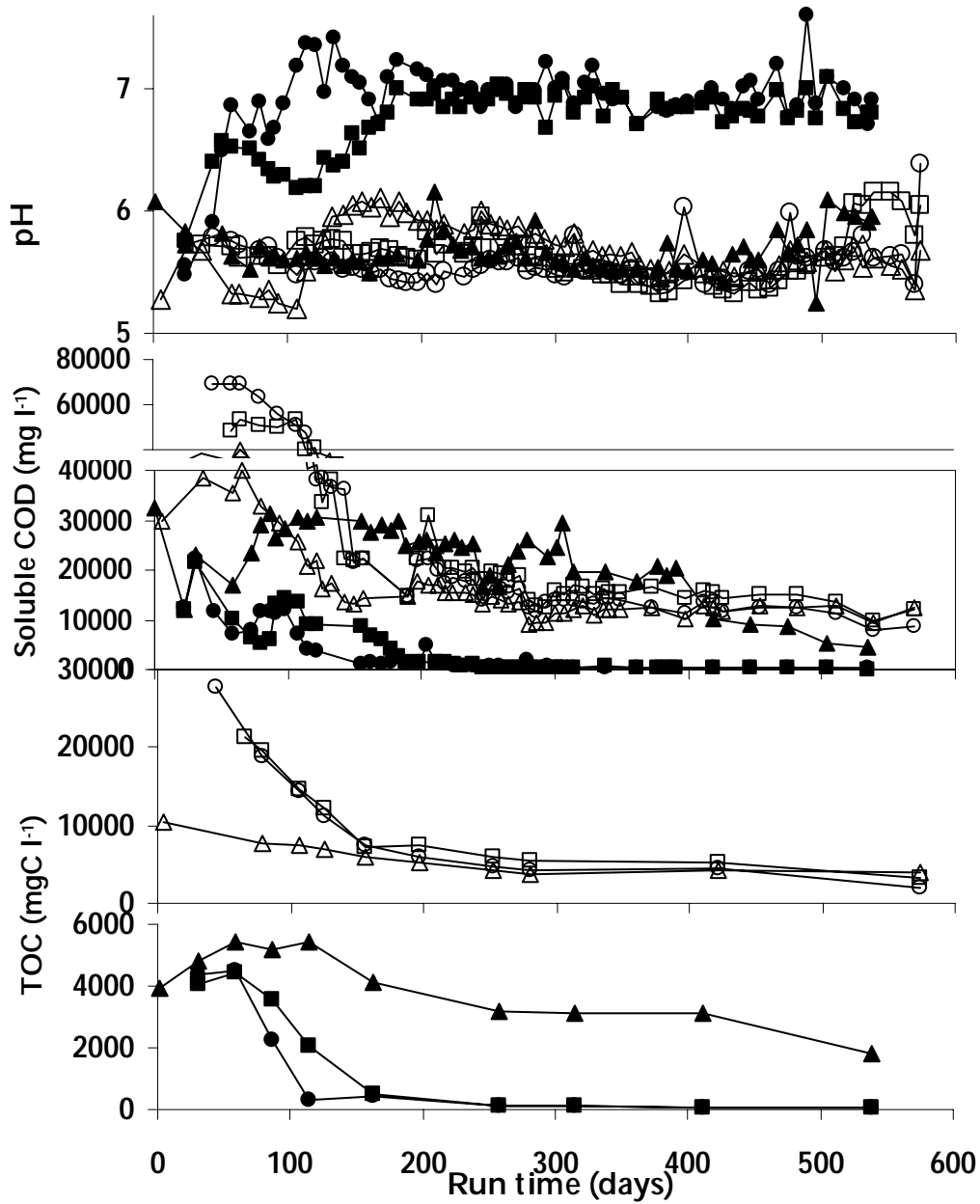

FIGURE 2   Effect of waste pre-wetting (L3 , L6 d), leachate recirculation and pH adjustment (L2 œ, L3, L5 › , L6) on methane content and cumulative methane production of the grey (L1' -L3) and the old landfill waste (L4 ' - L6) during the lysimeter experiment.

The effects of landfill procedure and waste type on leachate characteristics are presented in Fig. 3 as pH, SCOD and TOC, in Table 8 as BOD_7, 1 BOD_7/SCOD, VFA-COD/SCOD, DOC/SCOD and NH_4-N/TKN, VFAs in Fig. 4. The cumulative amounts of SCOD and NH_4-N released in the leachate against the L/S ratio are presented in Figs. 5 and 6, NH_4-N and TKN concentration in Fig. 7, total and specific release of COD into the leachate and biogas as methane as well as the specific release of NH_4-N and TKN in table 9. Generally, more COD and NH_4-N were released into the leachate from the grey waste per TS than from the MSW and the COD was released from the MSW into the biogas as a result of higher methanogenic activity. More COD was removed from the MSW without pre-wetting (L4 and L5 in Table 9) compared to the pre-wetted MSW.
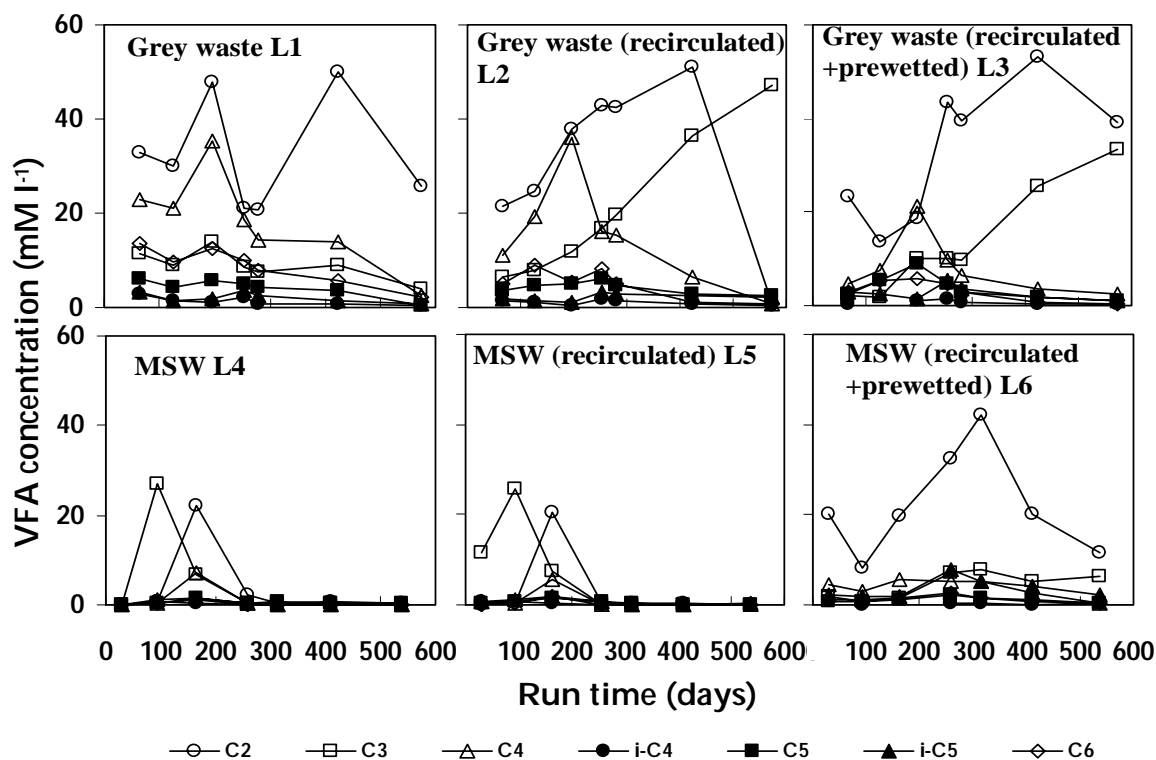
FIGURE 3   Effect of waste pre-wetting (L3   , L6 d) and leachate recirculation and pH adjustment (L2 œ , L3, L5 › , L6) on leachate pH, SCOD and TOC of the grey (L1 ’ -L3) and the old landfill waste (L4 ‘   - L6) during the lysimeter experiment.

TABLE 7   Effect of pre-wetting (L3, L6) and leachate recirculation (L2, L3, L5, L6) on total COD released via biogas and leachate, cumulative methane production, and on specific methane yield of the grey and old waste during the lysimeter experiment.

Lysimeter	Total released COD (gO$_2$)	Cumulative CH$_4$ production		Specific CH$_4$ yield		
		(l)	Proportion of released COD , (%)	(ml g$_{waste}^{-1}$)	(ml gTS^{-1})	(ml gVS^{-1})
Grey waste:						
L1 No recirculation	325	2.8	2.3	0.60	0.95	1.6
L2 Recirculation (pH7)	250	15	15	3.2	5.2	5.5
L3 Prewetted + recirculation (pH8)	408	5.5	3.5	1.3	2.1	3.5
Old landfilled waste:						
L4 No recirculation	1001	360	96	48	110	210
L5 Recirculation (pH7)	825	300	96	40	94	180
L6 Prewetted + recirculation (pH8)	513	74	38	9.5	22	42

In the beginning of the experiment, acidic leachate (pH below 6) and high concentrations of soluble organic matter (high SCOD, TOC, DOC/TOC and VFAs) was produced from all six lysimeters into leachates. However, in the leachate of the landfilled MSW (L4 and L5) pH increased to around 7 after the initial stages with the co-occurrence of high methanogenic activity and low concentrations of soluble organic matter (SCOD, TOC, DOC/TOC and VFAs), but the leachate of the pre-wetted MSW lysimeter (L6) remained acidic (between 5.5 and 6.0 and SCOD from 25 to 5 gCOD l^{-1}) throughout the experiment containing half of SCOD as VFA-COD. All the grey waste leachates were acidic for most of the time (pH 5.4 to 5.8 and SCOD from approx. 60 to around 15 gCOD l^{-1} in the end). Similar pattern with an equally high initial level of COD has been found with unsorted MSW in most of comparable studies of young landfill leachate (reviewed by e.g. Lema *et al.* 1988). The SCOD was at the latter part of the experiment mainly in form VFA-COD as shown by the high VFA-COD/SCOD ratio indicating of high level of waste hydrolysis and acidogenesis. In the final stages of the experiment (from day 520 onwards), the pH rose above 6 in grey waste lysimeter with recirculation only (L2) with the co-occurrence of increased methane production, indicating alleviated acidity and the initiation of methanogenic activity. Therefore, the launch methanogenesis is required to obtain leachates with low organic material concentrations and the alkali supplementation along with leachate recirculation was insufficient in the short term to balance the production (acidification) and consumption (methanogenesis) of VFA's as intermediate products, once the

acids had accumulated and the pH was at a low level. Therefore, it seems that some other strategies – such as the introduction of methanogenic inoculum, e.g. digested sludge, into the waste (reviewed by e.g. Barlaz et al. 1990) or recirculation of leachates containing methanogens - are needed to obtain increased methanogenesis, which could also provide better quality effluent. Another approach could be to recirculate the leachate containing acids to a methanogenic part of the landfill where it can be methanised (Reinhart & Townsend 1998).

FIGURE 4   Effect of waste pre-wetting (L3   , L6 d) and leachate recirculation and pH adjustment (L2 œ, L3, L5 › , L6) on leachate VFAs (acetic (C2), propionic (C3), butyric (C4), iso-butyric (i-C4), valeric (C5), iso-valeric (i-C5) and caproic acid (C6)) of the grey (L1 ' -L3) and the old landfill waste (L4 ' -L6) during the lysimeter experiment.

At the start, the grey waste leachate had high concentration of acetate, propionate and butyrate (L1, L2 and L3 in Fig. 4). Acetate and butyrate are commonly the main VFAs in landfill leachates at the early stages after landfilling of waste as suggested by the previous laboratory and full-scale studies (e.g. Rees 1980; Chugh et al. 1998; Pohland & Kim 1999). After day 200, acetate began to accumulate peaking on day 422 to approx. 70 mM l^{-1} in all the grey waste leachates, after which all the VFA concentrations decreased, except propionate in the recirculated leachates, which was 63 (L2) and 44 mM l^{-1} (L3) at the end. The high butyrate concentration might arise from inhibitory effect of propionate on butyrate oxidation (James et al. 1998). The pre-wetted MSW leachate (L6) had a high acetate concentration (20 mM l^{-1}). This suggests of

reduced acetoclastic methanogenic activity. Thus, it seems that the rapid addition of moisture as in waste pre-wetting (present study) may result in a low pH and the accumulation of acids and thus retard the methanation and hydrolysis of the waste, which in turn may sour the leachate in landfills. Therefore rapid saturation of waste should be avoided when applying leachate recirculation to the methanogenic part of the landfill. Methanogenic and acidogenic spaces usually coexist in landfill body and thus the high VFA concentrations would be consumed in the methanogenic waste body, as found in the pre-wetted lysimeter in the present study. Furthermore, in case of acidified waste in the landfill body, the halting of leachate recirculation and/or the addition of water might result in a swifter revival of methanation.

TABLE 8   Effect of waste pre-wetting (L3, L6) and leachate recirculation and pH adjustment (L2, L3, L5, L6) on leachate BOD_7 BOD_7/SCOD, VFA-COD/SCOD, DOC/SCOD and NH_4-N/TKN of the grey (L1-L3) and the old landfill waste (L4-L6) at the initial stages, in the middle and at the end of the lysimeter experiment.

| Lysimeter	Run time (days)	BOD_7 (g l^{-1})	BOD_7/SCOD	VFA-COD/SCOD	DOC/SCOD	NH_4-N/TKN
L1 No recirculation	44 - 93	54	0.78	0.19	0.29	0.70
	252-336	10	0.73	0.79	0.28	0.82
	422-510	8	0.70	1.0	0.31	0.76
L2 Recirculation (pH7)	44 - 93	42	0.87	0.12	0.30	0.69
	252-336	11	0.73	1.0	0.29	0.82
	422-510	10	0.74	0.73	0.33	0.76
L3 Prewetted + recirculation (pH8)	44 - 93	29	0.82	0.11	0.28	0.66
	252-336	10	0.90	0.99	0.27	0.75
	422-510	9.9	0.77	0.78	0.33	0.97
L4, No recirculation	23-92	12	0.97	0.32	0.14	0.66
	162-272	0.048	0.10	0.96	0.26	0.76
	391-475	0.068	0.13	0.76	0.37	0.09
L5 Recirculation (pH7)	23-58	7.5	0.62	0.08	0.14	0.70
	162-272	0.05	0.11	0.79	0.26	0.29
	391-475	0.02	0.08	0.17	0.28	0.47
L6 Prewetted + recirculation (pH8)	23-58	9.2	0.75	0.13	0.11	0.70
	162-272	7.6	0.32	0.51	0.19	0.68
	391-475	6.1	0.71	0.46	0.28	0.03
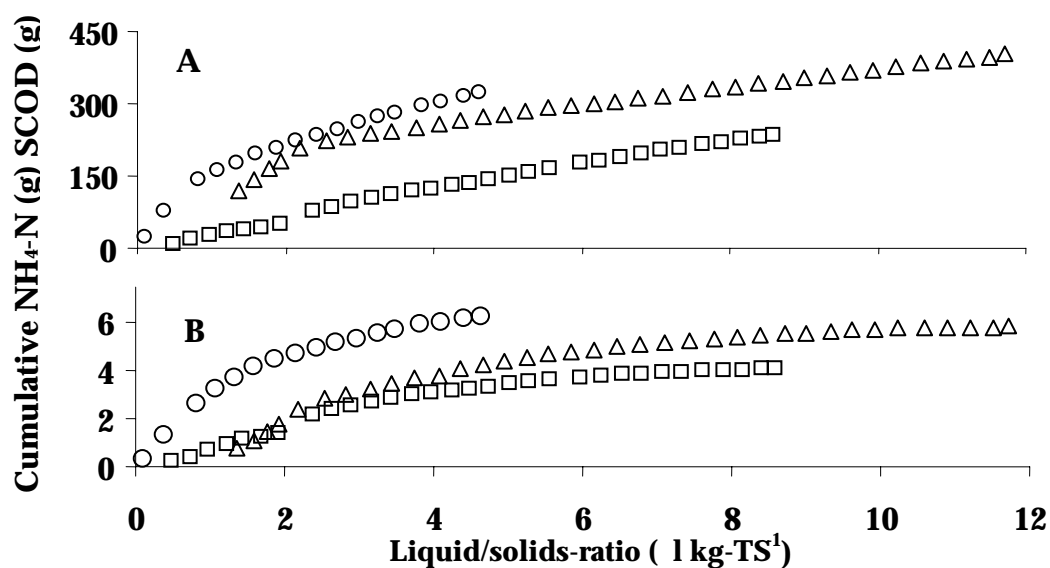
FIGURE 5 Relation to L/S ratio of cumulative release of SCOD (a) and NH4-N (b) into leachate from grey waste (L1 ˙ -L3f) with waste pre-wetting (L3 f) and leachate recirculation and pH adjustment (L2 œ) during the lysimeter experiment.

TABLE 9 Effect of pre-wetting (L3, L6) and leachate recirculation (L2, L3, L5, L6) on total COD released via biogas and leachate, cumulative methane production, and specific methane yield of grey and old landfill waste during the lysimeter experiment.

Operation	Total released COD (gO$_2$ kgTS^{-1})	Released as CH$_4$-COD (gO$_2$ kgTS^{-1})	Released as leachate-COD (gO$_2$ kgTS^{-1})	Released as leachate-NH$_4$-N (gN kgTS^{-1})	Released as leachate-TKN (gN kgTS^{-1})	Eluted COD after the experiment (gO$_2$ kgTS^{-1})
Grey waste:						
L1 No recirculation	111	2.5	109	2.1	2.7	0.61
L2 Recirculation (pH7)	94	13.7	80.6	1.4	2.7	0.69
L3 Prewetted + recirculation (pH8)	161	5.6	155	2.2	3.2	0.36
Landfilled MSW:						
L4 No recirculation	314	302	12.2	0.09	0.26	0.19
L5 Recirculation (pH7)	259	249	9.7	0.13	0.20	0.21
L6 Prewetted + recirculation (pH8)	155	59.1	95.7	0.24	0.57	0.22
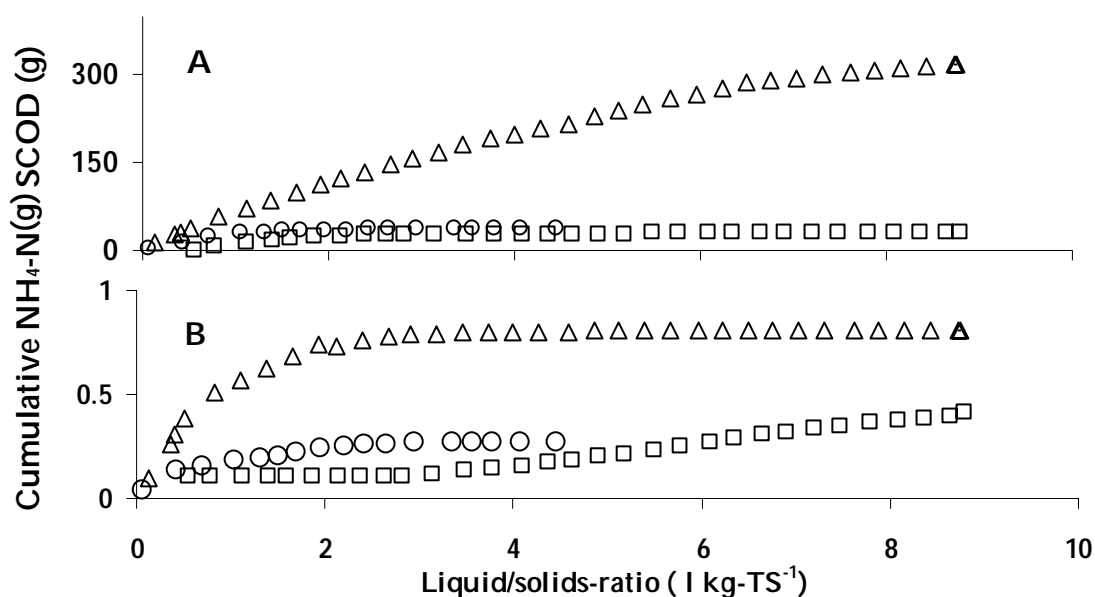
FIGURE 6   Relation to L/S ratio of cumulative release of SCOD (a) and NH₄-N (b) into leachate from the old landfill waste (L4 ' -L6f) with waste pre-wetting (L6) and leachate recirculation and pH adjustment (L5 œ) during the lysimeter experiment.

Both the NH₄-N and TKN concentrations from all the grey waste lysimeters were significantly higher than these in the MSW lysimeters (Fig. 7), which could indicate that most of the nitrogen in the MSW had been released during the 10 years it had been landfilled, because threefold TKN content (around 2 gN l⁻¹) has been reported for unsorted MSW landfill samples (Ham et al. 1993). The NH₄-N concentrations in the grey waste leachate were at the start 820-1500 mgN l⁻¹ and at the end around 100 mgN l⁻¹. The high level of nitrogen in the leachates apparently originated from easily degradable organic waste materials, e.g. proteins from residual putrescible waste fractions. Similarly, high leachate nitrogen concentrations (above 1 gNH₄-N l⁻¹) have been reported in both laboratory lysimeter (Ehrig 1989) and full-scale landfill studies (Reinhart & Al-Yousfi 1996) for unsorted MSW. In the MSW leachates NH₄-N fell from its initial higher values (150-200 mgN l⁻¹; highest in the pre-wetted lysimeter) to 10-40 mgN l⁻¹. The high NH₄-N in the pre-wetted lysimeter leachates could be due the increased flow of water through the interstices of waste particles thereby releasing the readily solubilised NH₄-N into the leachate. The longer residence time of the leachate in the pre-wetted lysimeter may have increased transfer of readily dissolved NH₄-N from the surface as well as interior of the MSW particles into the leachate. Hence, rapid moisture elevation combined with leachate recirculation increases the rate of dissolution of ammonium into the leachate and may be a method that can be applied to shorten the period of high ammonium concentration in the leachates from older, e.g. closed landfills.
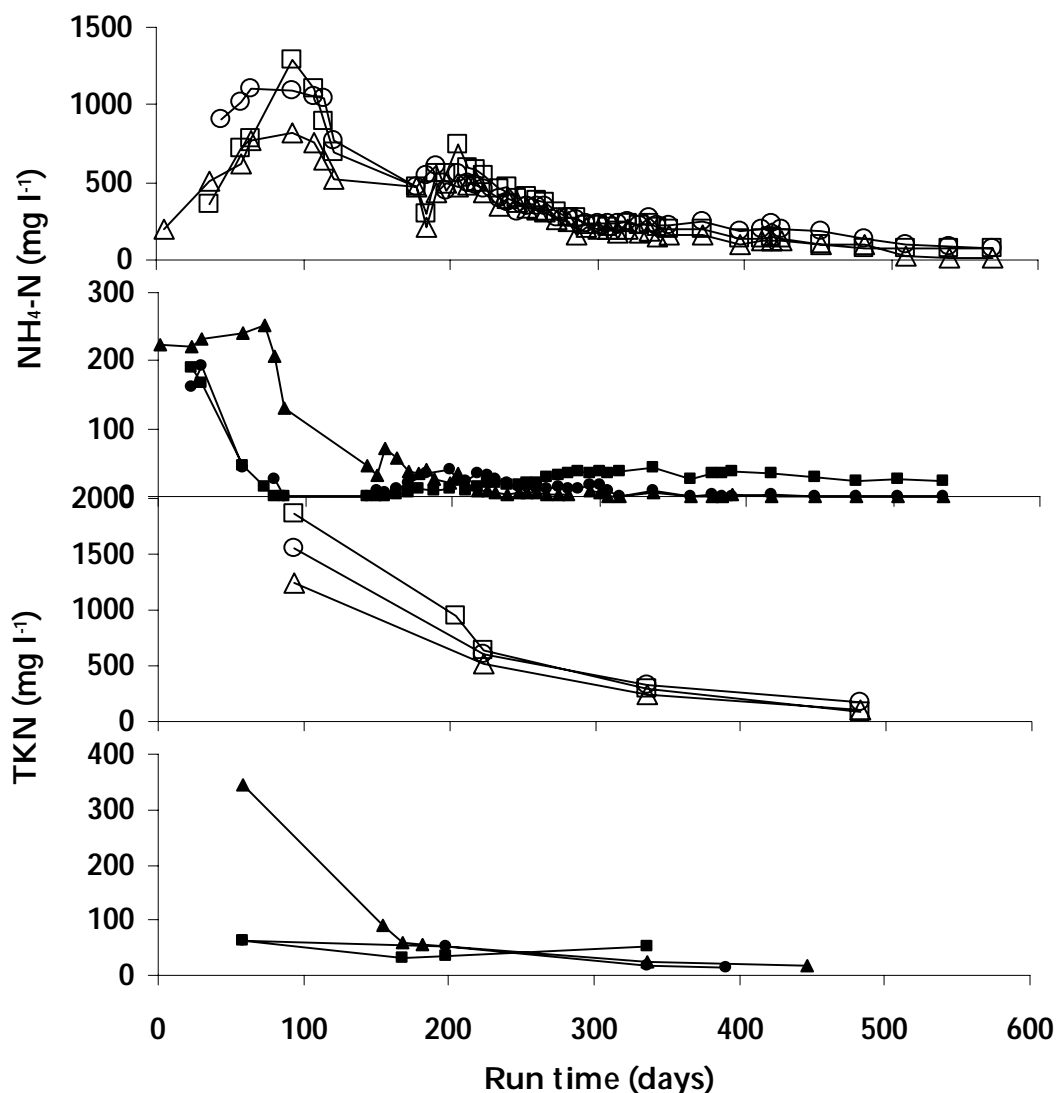
FIGURE 7   Effect of waste pre-wetting (L3f , L6d) and leachate recirculation and pH
adjustment (L2œ, L3, L5› , L6) on leachate NH₄-N and TKN of the grey (L1' -
L3) and the old landfill waste (L4' -L6) during the lysimeter experiment.

Most of the NH₄-N was produced from grey waste in the control lysimeter at low L/S ratios (L/S<2) suggesting of the rate of hydrolysis had a greater limiting effect on the release of ammonium nitrogen in the leachate than the rate of dissolution (after L/S ratio of 1). With the MSW, most of the NH₄-N was released into the leachate with a L/S ratio < 2. The rate of NH₄-N release accelerated in the MSW with recirculation at the higher L/S ratios indicating possible suppressive interaction by methanogens on proteolytic activity (Chen et al. 2000). Previously, in laboratory lysimeters with unsorted MSW, an L/S ratio between 5 and 10 was needed to decrease the leachate NH₄-N concentration from 400 mgN l⁻¹ to below 10 mgN l⁻¹ (Munasinghe 1997). A L/S ratio of 1 is equal to 30 years in the local landfill conditions without leachate recirculation if surface runoff and evapotranspiration are subtracted from the amount of precipitation, whereas with leachate recirculation in a 1:1 ratio to precipitation, the time period required is 15 years. In the local conditions, it

would take about 120 years before most, i.e. approx. 70 %, of the total released nitrogen leached from grey waste where no additional water is added. However, with application of an active bioreactor strategy including e.g. initial wetting and leachate collection and recirculation, the L/S ratio would be closer to the one applied in the present laboratory experiment. On the other hand, in landfill practice, the rate of water infiltration applied during leachate addition is limited e.g. by the permeability of water into the waste body (Reinhart & Townsend 1998).

4.4.2 Effect of waste aeration on methane and leachate emission potential

The effect of aerobic treatment on the methane and water pollution potential of various source-separated waste fractions was studied in lysimeters at 20ºC. The PFMSW, CPFMSW, grey waste, and the lysimeter landfilled grey waste and the lysimeter landfilled MSW samples were aerated continuously at an airflow rate between 50 and 30 l d^{-1} kg^{-1} of waste. The 51-day waste aeration decreased the cellulose content in all the wastes except CPFMSW (Fig. 8). Aerobic treatment of 51 days decreased the cellulose content of PFMSW by 70 %. In all samples methane production started without a significant lag. In a study similar to present one, a 75 % reduction in VS was obtained in 41-day aerobic treatment of PFMSW, in laboratory reactors using sawdust as the bulking agent (Koenig & Tao 1996).
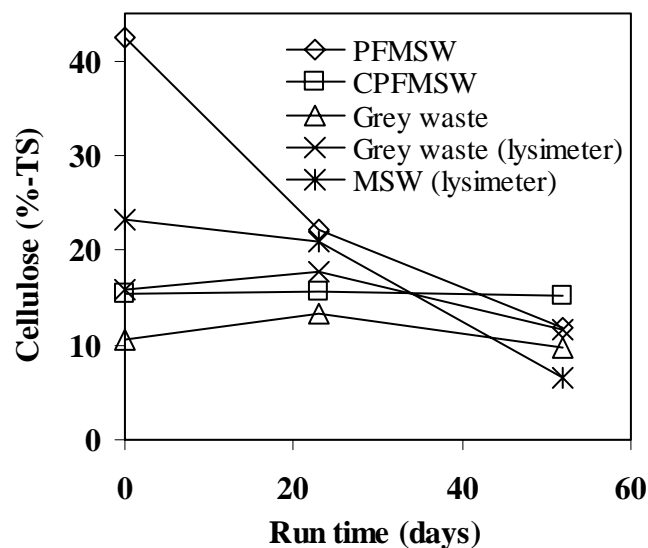
FIGURE 8   Cellulose content of the waste samples during aerobic lysimeter test.

A biochemical methane potential (BMP; incubation period 110 d) test was carried out on all five wastes and on samples from the aerobic lysimeters on days 23 and 52 (Fig. 9 and Table 10). The methane yield of PFMSW (410 m^3 tTS^{-1}) was approx. tenfold compared to that in the residual grey waste. Similar

methane yields were obtained also with a continuously stirred tank reactor (476 m^3 tTS^{-1} (calculated by authors); Cecchi et al. 1992) at 35°C with HRT of 50 days in the source-separated PFMSW suggesting that a significant reduction in methane production would be achieved by removal of this fraction (reviewed by Komilis et al. 1999b). Residual grey waste resulting from source-separation has a methane potential of 46 m^3 tTS^{-1} (this study) to 100 m^3 tTS^{-1} (II), a yield up to 526 m^3 tTS^{-1} of methane was obtained by anaerobic treatment of grey waste in a two-stage concept (with HRT of 4.3 d at 65°C followed by 14.2 d at 55°C) (Scherer et al. 2000). The level of methane yield, however, is dependant on the methods of separation and mechanical pre-treatment used.

Aeration generally decreased the maximum methane production rate, whereas in both grey waste samples aeration for 23 days increased the rate by 28 %, but 51 days of aeration lowered the rate in the grey waste and in the lysimeter grey waste by 85 % and 74 %, respectively, compared to initial values, respectively suggesting a low rate of degradation of components of the grey waste. Therefore, when applying controlled landfill aeration, preferably much longer than 51 days would be needed to obtain a high reduction in the methane potential of landfilled waste. A long-term treatment (up to 32 weeks) was needed to gain high degree of stabilisation, i.e. a biogas (CH_4+CO_2) potential of less than 20 m^3 tTS^{-1}, in a study with mechanically pre-treated MSW (Höring et al. 1999). The fact that 23-day aeration increased the specific methane yield of the grey waste samples studied here could indicate an increase in the availability of organic polymers for methanogenesis aided by aerobic hydrolysis of the material. Enhanced hydrolysis achieved by the pre-composting of MSW resulted in higher methane production in anaerobic digestion than obtained with non-precomposted MSW (Mata-Alvarez et al. 1993). Therefore, a short-term aeration could possibly be used to enhance the degradation of the grey waste into methane.

The low methane potential (26 m^3 tTS^{-1}) of the landfilled MSW samples after the long-term anaerobic laboratory lysimeter experiment (Jokela et al. 2001) indicated the limited potential of this waste type for further anaerobic degradation after the 51-day aeration. In the present BMP studies, the use of a small maximum particle size (2 mm) and optimal inoculation enhanced methanation. In landfills the degradation rate and potential of MSW would probably be lower owing to limitations, such as the lower temperature in landfills and the larger particle size of the MSW.
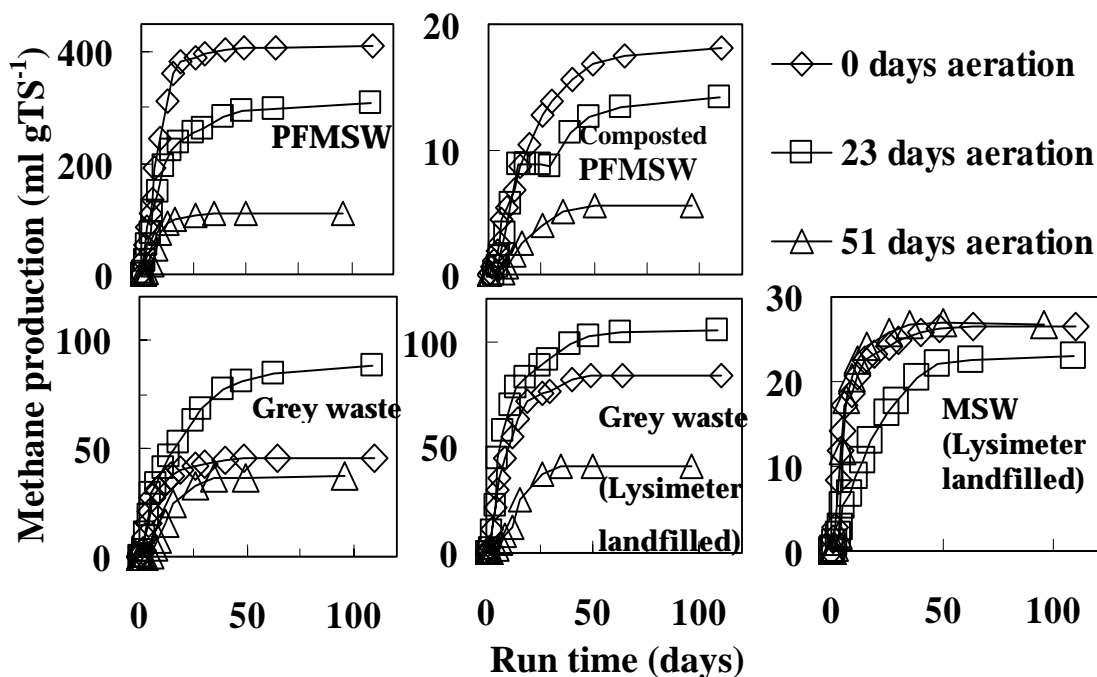
FIGURE 9   Cumulative methane production from PFMSW, CPFMSW, grey waste, lysimeter landfilled grey waste and MSW in the BMP test at 35\C with added methanogenic inoculum with samples obtained on days 0, 23 and 51 from aerobic treatment.

TABLE 10   Specific methane yields from PFMSW, CPFMSW, grey waste as well as lysimeter landfilled grey waste and MSW in the BMP with 0, 23 and 51 day samples of aerobic treatment (standard deviation in parenthesis).

Sample	Specific methane yield					
	Day 0		Day 23		Day 51	
	$(mlCH_4$ $gTS^{-1})$	$(mlCH_4$ $gVS^{-1})$	$(mlCH_4$ $gTS^{-1})$	$(mlCH_4$ $gVS^{-1})$	$(mlCH_4$ $gTS^{-1})$	$(mlCH_4$ $gVS^{-1})$
PFMSW	410(42)	527	310(30)	400	109(73)	165
CPFMSW	18(8)	41	14(0.8)	40	5.6(0.5)	15
Grey waste	46(6)	76	89(7)	130	37(0.2)	65
Lysimeter landfilled grey waste	84(14)	140	105(4)	147	41(22)	69
Lysimeter landfilled MSW	26(5)	60	23(4)	51	27(2)	65

The leaching of soluble COD and NH_4-N was studied by means of the elution test (Fig. 10) from the five waste materials and from the samples obtained from the aerobic lysimeter experiment on days 23 and 51. On day 23 of aeration, a pronounced decrease in elutable NH_4-N was observed in the grey waste samples and the 51 days aeration increased the elutable NH_4-N concentration of PFMSW up to 7.9 gNH_4-N $kgTS^{-1}$. The ammonium emission potential of the PFMSW was significantly higher than that of either grey waste or CPFMSW. The PFMSW also seems to be prone to rapid nitrogen solubilisation as a result

of aerobic/anaerobic degradation. Previously, on the basis of the nitrogen emission potential of MSW derived using different methods has been estimated to be between 1.4 and 3.4 gNH_4-N $kgTS^{-1}$ and the TKN content between 2.0 and 4.0 g $kgTS^{-1}$ (Kruempelbeck & Ehrig 1999). However, the shaking test yields merely rough estimates of the possible long-term emission potential of landfilled MSW, and the long-term nitrogen emission potential of landfill cannot be derived from tests carried out in laboratory conditions, as previously concluded (Heasman 1997).
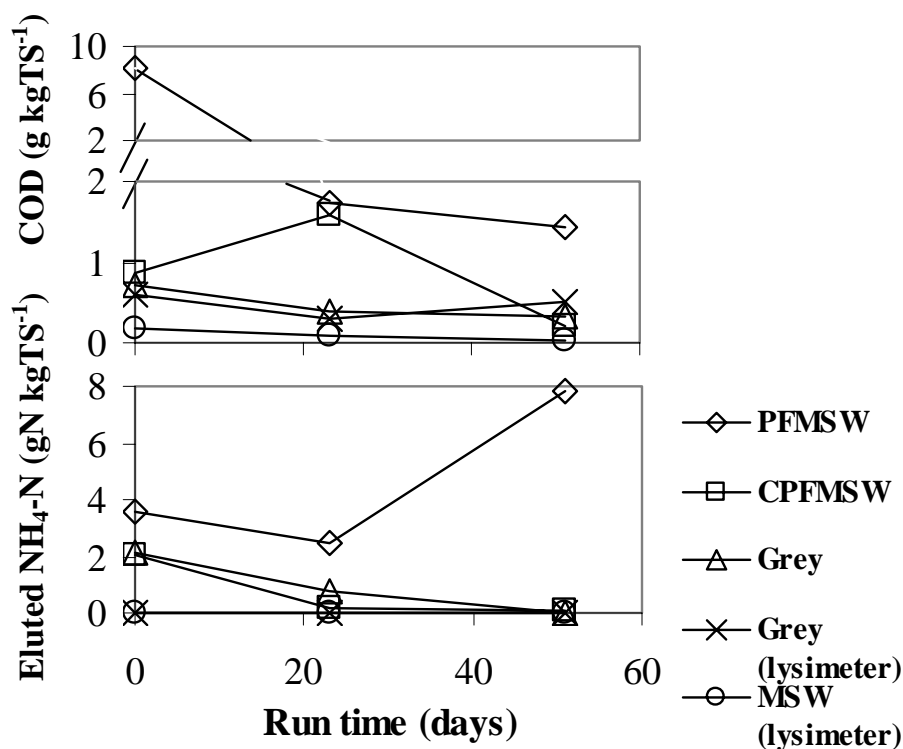
FIGURE 10   Leaching of soluble COD and NH_4-N during the elution test from PFMSW, CPFMSW, grey waste as well as lysimeter landfilled grey waste and landfilled MSW and with 23 and 51 day samples of aerobic treatment.

4.5 Biological nitrogen removal from municipal landfill leachate

4.5.1 Low-cost nitrification in biofilters

Nitrification of landfill leachate was studied in an upflow filter with crushed brick as the filter material in both the laboratory- (UF) (Fig. 11) and on-site pilot study (pilot UF) (Fig. 12) as well as in the laboratory DF with mature compost as the filter material (Fig. 13) and with the laboratory-scale SCBP as a reference for the laboratory filters (Fig. 14). In the laboratory nitrification filters, start-up of nitrification took less than three weeks and over 90 % nitrification of leachate (NH_4-N between 60 and 170 mgN/l, COD between 230 and 1300 mg/l) was

obtained with loading rates between 100 and 130 mgNH₄-N/I d at 25°C. Ammonium concentrations in all biofilter effluents were usually below the detection limit. The laboratory and pilot studies show that filters employing low cost waste materials as filter media provide for the rapid and stable nitrification of landfill leachate. Previously, in an on-site biofilter study ammonia removal from municipal landfill leachate above 90 % with an ammonia loading rate of 70 mgN I^{-1} d^{-1} was reported using a trickling filter (commercial carrier media) at 16°C (Knox 1984). In practice, nitrification might be more feasible in an upflow mode filter due to longer HRT, which could compensate the slower biological reactions at low temperatures. Crushed concrete demolition waste could probably also be used as a filter medium, whereas the applicability of compost material could be limited owing the possible flotation of this material in the upflow mode.
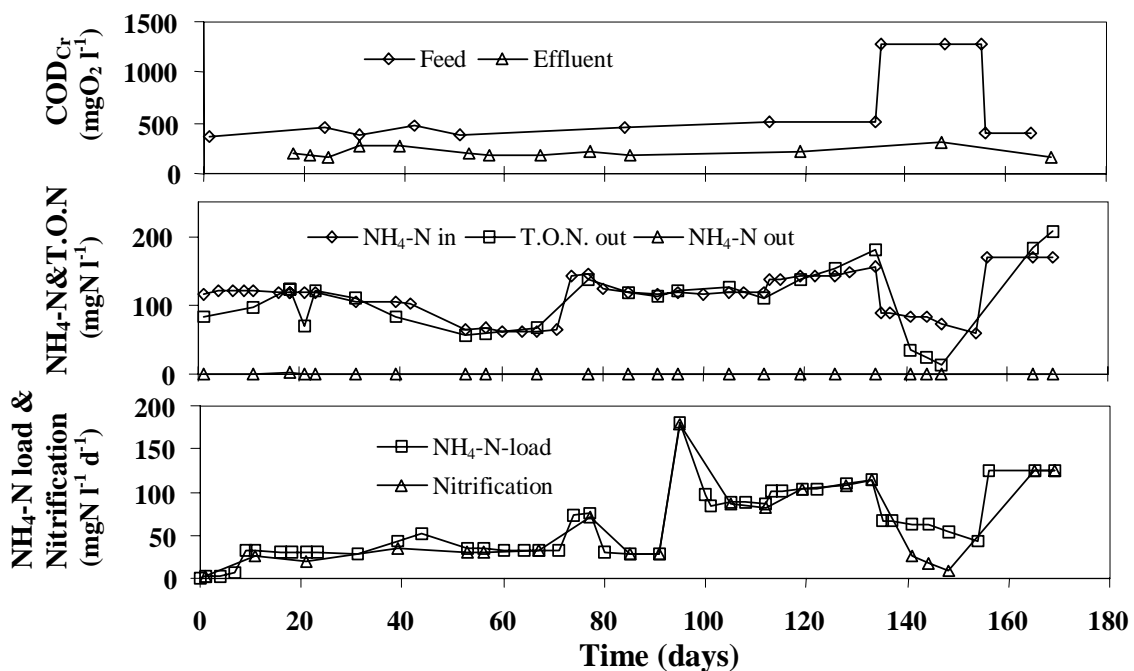
FIGURE 11   Feed COD and NH₄-N, effluent COD, NH₄-N and T.O.N., and NH₄-N load and nitrification efficiency in a laboratory experiment using an upflow filter with crushed brick as the filter medium to treat municipal landfill leachate. HRT: semicontinuous feeding, 3.8 d, and 1.4 d between days 1-8, 9-91, and 91-end, respectively.

In the pilot study upflow filter, above 95 % nitrification of landfill leachate (NH₄-N between 160 and 270 mgN/I, COD between 1300 and 1600 mg/I) was obtained in low temperature conditions (at 5°C and 10°C) with a loading rate of approximately 50 mgNH₄-N I^{-1} d^{-1}. Similarly, in a laboratory SCBP study (Hoilijoki et al. 2000) complete ammonium removal was obtained at 5°C and

10°C with loading rates of 25 and 50 mgNH₄-N g l⁻¹ d⁻¹, respectively. Given the similar level of loading rates applied on these two processes, the use of waste material as biofilter media for full-scale application might also be economically attractive.
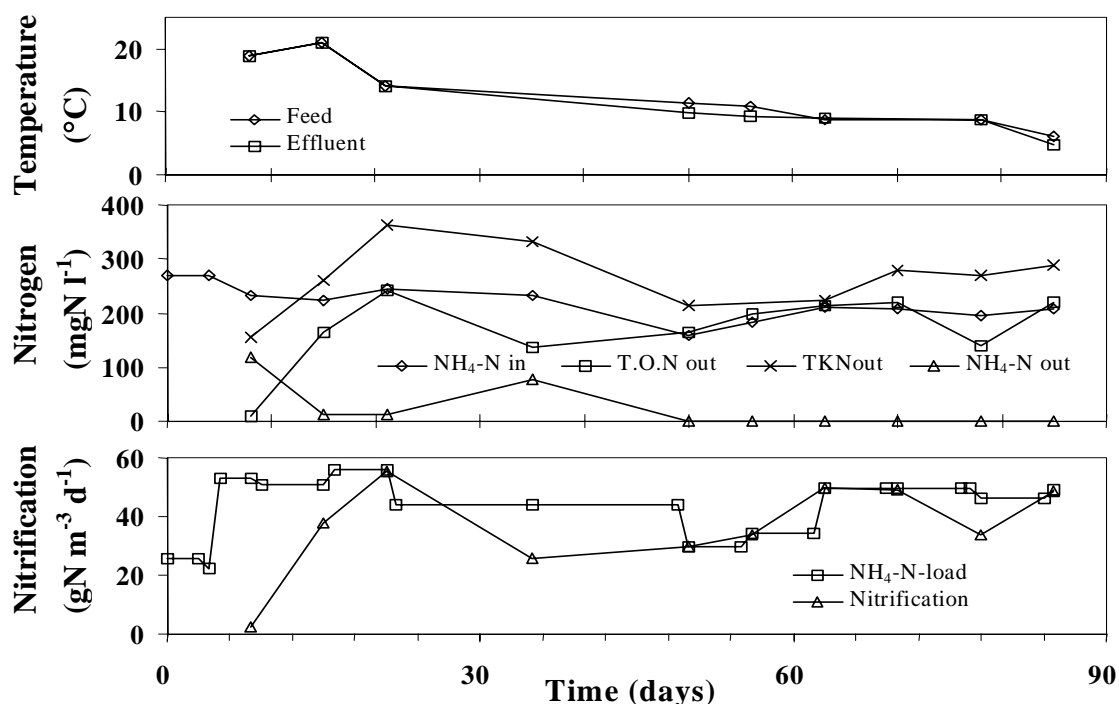

FIGURE 12   Temperature, feed NH₄-N and effluent NH₄-N, TKN and T.O.N. and NH₄-N load and nitrification efficiency during nitrification of municipal landfill leachate in on-site pilot experiment in an upflow filter with crushed brick used as the filter medium. HRT: semicontinuous feeding, 2.2 d, 2.6 d, and 2.1 d between days 1-3, 4-20, 21-60, and 61-85, respectively.

The exceptionally high COD in leachate feed dropped the nitrification efficiency in UF to less than 20 % and caused inhibition of nitrification in the SCBP. This was probably caused by the competitive advantage of heterotrophs over nitrifiers. Onay & Pohland (1998) also concluded that the anaerobic treatment of leachate prior to in-situ nitrification resulted in a higher level of nitrification. Therefore, when applying in-situ nitrogen removal, leachate COD and ammonia concentrations and their changes need to be taken into account, especially in landfills containing leachate with varying characteristics and age.
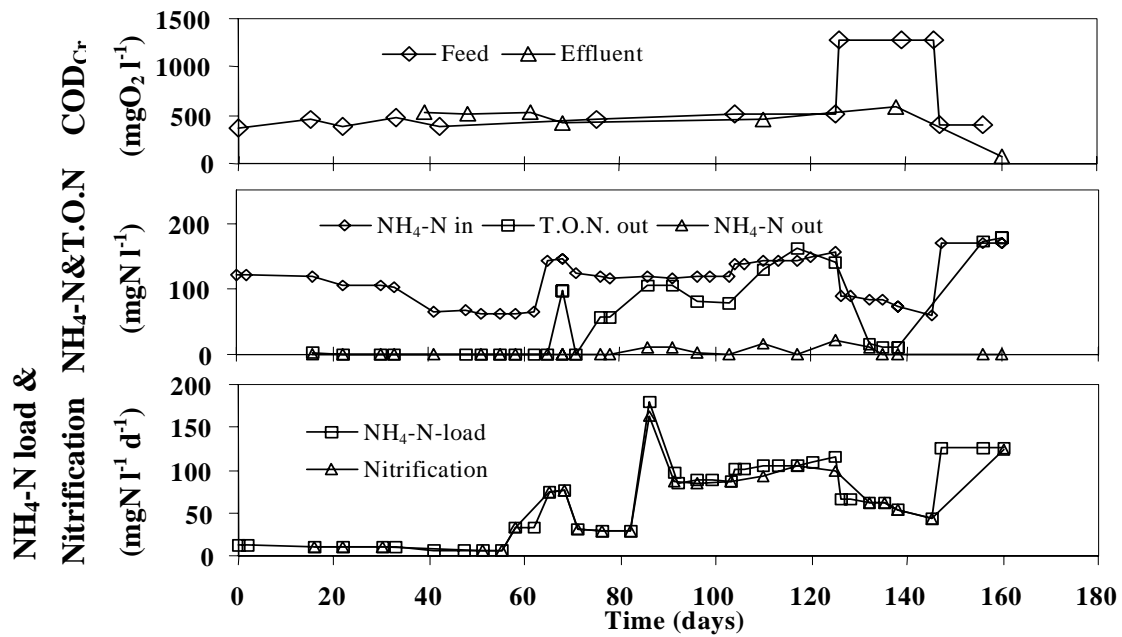
FIGURE 13   Feed COD, NH₄-N and effluent NH₄-N, and T.O.N. and NH₄-N load and nitrification efficiency during nitrification of municipal landfill leachate in a continuous laboratory downflow filter with mature compost as the filter media. HRT: 9.6 d, 5.1 d and 7.3 d between days 1-58, 59-91, and 91-160, respectively.
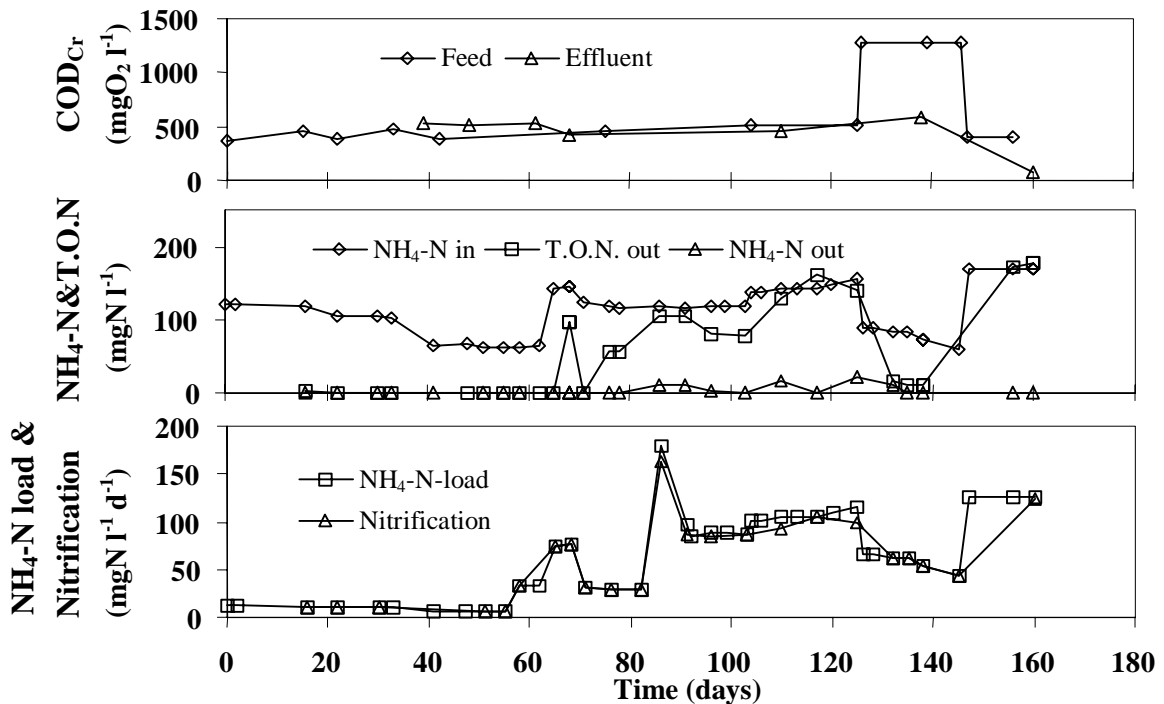

FIGURE 14   Feed COD and NH₄-N and effluent COD, NH₄-N and T.O.N. and NH₄-N load and nitrification efficiency during nitrification of municipal landfill leachate in laboratory suspended carrier biofilm process (SCBP). HRT: 0.6 d, 2.6 d, 5.1 d, 2.4 d, 7.3 d and 4.6 d between days 1-13, 14-108, 109-147, 148-169, 170-216, and 217-254, respectively.

4.5.2 In-situ denitrification

Denitrification of nitrified leachate was studied in the laboratory in a landfill waste column by feeding semi continuously nitrified leachate from SCBP into the column. At the beginning some T.O.N (5.5 mgN l^{-1}) was found in the effluent, while after 40 days it was no longer detected (Fig. 10). Effluent COD was higher than feed COD except in the last sample. Initially, the loading rates were between 0.36 and 0.40 mgT.O.N l^{-1} d^{-1} (until day 185) and 0.6 mgT.O.N l^{-1} d^{-1} (until the end). The nitrified landfill leachate can be denitrified in a landfill body that contains old methanogenic waste material with the loading rate of 3.8 gT.O.N t-TS$^{-1}_{waste}$ d^{-1}. In a laboratory batch study by Burton & Watson-Craik (1999), addition of nitrate (500-1000 mgNO$_3$-N l^{-1}, added as KNO$_3$-N) into methanogenic landfilled MSW resulted a complete denitrification with the rate equivalent approx. to 470 and 1100 gNO$_3$-N t-TS$^{-1}_{waste}$ d^{-1} (calculated by authors). However, this resulted delayed onset of methanogenesis and reduced quantities of methane produced. In their study the added initial nitrate load was about hundred times higher than in our study, which probably explains the difference in the effects on methanogenesis.

The amount of ammonia in the denitrified leachate showed a slightly increasing trend in the course of the run (Fig. 10). This may have originated from the landfill material used in this column. On the other hand, the high COD/NO$_3$-N ratio of the leachate may promote the reduction of nitrate of leachate to ammonia (Akunna et al. 1992), and as the COD/NO$_3$-N ratio inside the column was not measured, the possibility that nitrate removal could have been due to reduction to ammonia in the column can not excluded. However, the few gas samples analysed showed the presence of nitrogen (data not shown) that supported the fact that denitrification was the major mechanism of nitrate removal. Apparently, the waste materials in the denitrification column contained sufficiently available organic material and nutrients (especially phosphorous) for denitrification during the run. On the other hand, during long-term operation of denitrifying landfill body, their sufficiency should also be considered.

The methane content of the gas produced and the cumulative amount of methane produced in the denitrification column and in the control column are presented in Fig. 16. This suggests that the loading rate of T.O.N to the methane-producing part of landfill can be at least 3.8 gN t-TS$^{-1}_{waste}$ d^{-1} without any adverse effect on methanogenic activity. This means that, for example, about 60 m^3 of nitrified leachate with 100 mgT.O.N l^{-1} could theoretically be recirculated daily in a 100 m^2 landfill area with a depth of 20 m (density 0.8 t m^{-3}). The maximum denitrification capacity of the landfill material was not studied in the present experiments, but would presumably be higher than the level used here as the results of Burton & Watson-Craik (1999) presented above suggest.

Denitrification could probably also be employed during colder periods as the temperature of the biologically active part of a landfill body is agreeable (4-26°C) even during colder periods (Ettala 1988; Airiola & Lakso 1996).
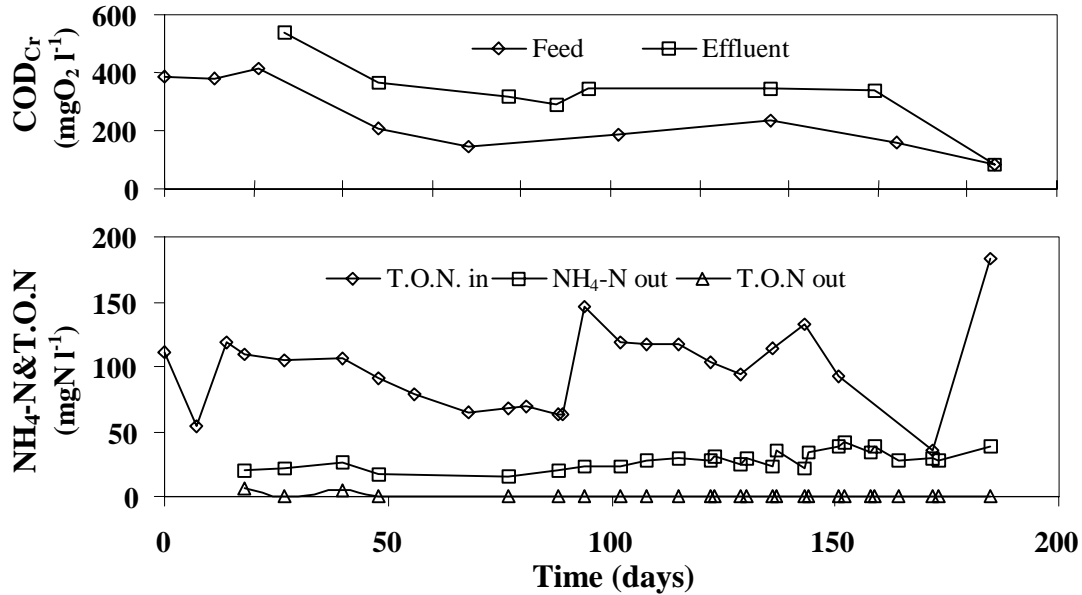

FIGURE 15   Feed COD and T.O.N and effluent T.O.N. and NH4-N in denitrifying waste column during denitrification of nitrified (suspended carrier biofilm process) municipal landfill leachate in the laboratory. The column contained horizons of approx. 10 and 40 years old municipal landfilled waste.

When applying denitrification in a landfill body by the recirculation of nitrified leachate, there are a number of technical matters that should be taken into account. These include design and selection of even distribution of leachate to avoid destabilising the waste body by flooding it (reviewed by Reinhart & Townsend 1998), a drainage clogging during the recirculation of leachate by increased its particulate content of leachate as well as the channel flow of leachate that may both cause an excessive head of leachate potentially causing failure of the bottom liner (reviewed by Reinhart & Townsend 1998) and the limitation of leachate recirculation by the low hydraulic conductivity of well-compacted landfill waste (approx. 10^{-5} – 10^{-8} m s^{-1} (reviewed by Saarela 1997)). Thus, only 0.1 – 90 m^3 d^{-1} of leachate can be recirculated on 100 m^2 of landfill site. Also, the parts of the landfill subject to leachate recirculation must be carefully selected as excessive addition of leachate by recirculation could retard the methanation of fresh waste (Jokela et al. 1999).
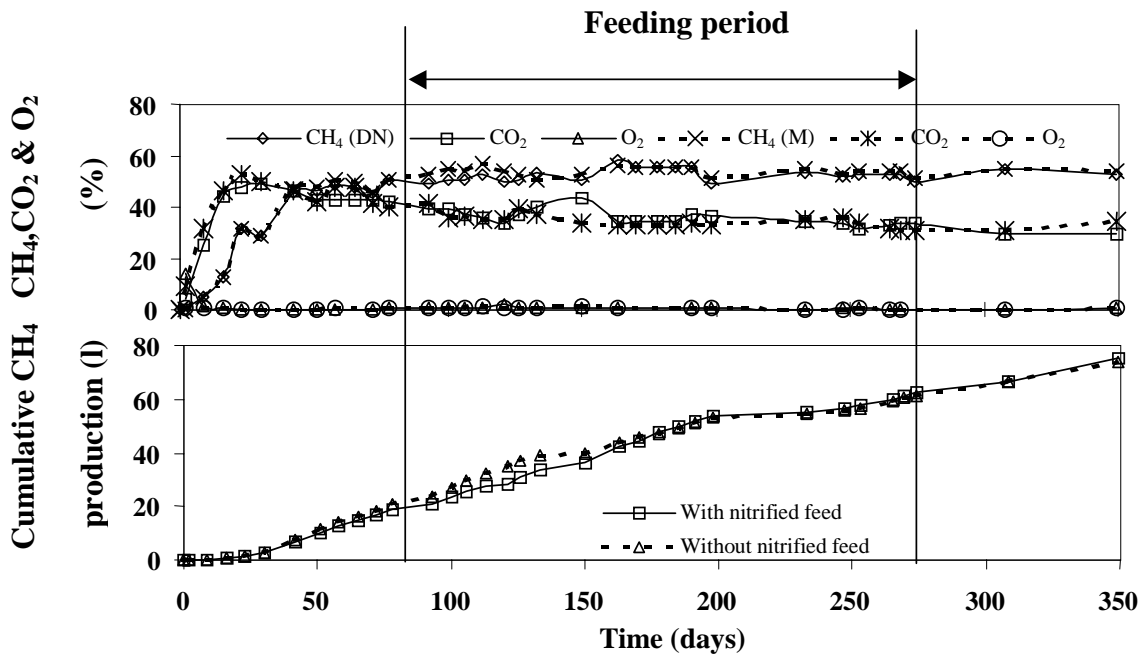
FIGURE 16   Methane, carbon dioxide (CO_2) and oxygen concentration of biogas and cumulative methane production in denitrifying (DN, solid line) and control (M, without leachate addition, dash line) waste column during denitrification of nitrified (suspended carrier biofilm process (SCBP)) municipal landfill leachate in the laboratory.   Both columns contained horizons of approx. 10 and 40 years old municipal landfilled waste.

5 CONCLUSIONS

The continuous disposal of MSW in landfills is causing methane emissions in the form of landfill gas and nitrogen emissions in the form of landfill leachate. A major reduction in greenhouse gases could be achieved by decreasing methane emissions originating out of the practices of waste management, especially the landfilling of waste. At present, the major proportion of nitrogen-containing MSW ends up in landfills; for example, in the EU countries around 2 million tonnes of nitrogen are disposed of in landfills annually. With present waste management practices nitrogen will continue to leach out of landfills for many decades to come. The short and long-term effects of landfilling of MSW in form of landfill gas and leachate emissions can, however, be controlled by landfill operation as well as by waste pre-treatment, if the characteristics of the MSW and the effects of waste pre-treatment on it as well as the biological and the physical-chemical parameters affecting its degradation during landfill operation are taken into account.

The grey waste produced after source-separation contains around 70 % w/w of biodegradable waste components that could under anaerobic conditions and in the presence of methanogenic microorganisms yield high amounts of methane (between 120 and 320 m^3 tTS^{-1}) and nitrogen (between 0.32 and 3.5 gNH_4-N $kgTS^{-1}_{added}$). The first-order rate constant of hydrolysis was in the range of 0.021 and 0.058 d^{-1}, which is around one-sixth of the values reported previously for source-separated putrescibles. The effective separation and biological treatment of putrescibles are thus important in reducing the environmental impact of waste disposal in landfills, since untreated PFMSW has a high methane (410 m^3 tTS^{-1}) yield and NH_4-N (3.6 $kgNH_4$-N tTS^{-1}) leaching potential. Long-term biological treatment, e.g. the combination of aerobic and anaerobic pre-treatment, is needed to reduce emission potentials, e.g. to ensure a biogas potential below 20 m^3 tTS^{-1} before final disposal to landfill. Controlled landfill aeration, preferably much longer than 51 days, could result in a high reduction in the methane potential of landfilled waste.

Various parameters tested in the laboratory indicated that waste degradation in the landfill could be affected. Low moisture conditions (i.e. below field capacity) can significantly suppress (old landfill waste) or prevent (grey waste) the methanation of landfilled waste, and therefore controlled access to water and/or leachate is recommended when an impermeable top cover is used on landfills. The landfill lysimeter studies showed that landfill operation procedures, in addition to moisture control, can drastically affect waste methanation and the characteristics of the leachate, and thus, an active bioreactor approach may prove to be a feasible method in landfill management. A high rate of methanogenic activity in landfilled MSW seems to promote waste hydrolysis by removing inhibitory intermediates and balancing the pH, whereas lowering the pH retards waste hydrolysis. Waste methanation removes COD from the waste body rapidly and thus leading to shorter operation times. This requires, however, a biogas collection and utilisation system to prevent the uncontrolled release of greenhouse gas into the atmosphere. The rapid addition of moisture to landfilled MSW by pre-wetting acidified waste inhibited methanogenesis thus causing a high and long-lasting COD load on the leachate. Leachate recirculation with pH adjustment to 8 with addition of bicarbonate buffer was insufficient to convert the waste from acidic to methanogenic. Landfilled grey waste produces significant amounts of methane (70-85 % of the total yield of MSW) or high strength sour leachate and over a long period if the waste is not rendered methanogenic by e.g. the addition of inoculum. On the basis of the L/S ratio applied, grey waste in landfill would produce a high COD and NH_4-N load in the leachate for at least 120 years, but with the controlled addition of water and leachate recirculation that period could be shortened to a couple of years. The release of ammonium from methanogenic MSW in landfills can be enhanced by raising the moisture level, e.g. by leachate recirculation, to obtain a L/S ratio between 3 and 4.

Nitrogen can be removed effectively from municipal landfill leachate by using a nitrifying upflow biofilter with waste material as a filter medium combined with subsequent denitrification of the nitrified leachate in the landfill body. Crushed waste brick can be used as a carrier material in upflow filters for nitrification of municipal landfill leachate with loading rates of 100-130 and 50 $mgNH_4$-N l^{-1} d^{-1} at 25ºC and at 5ºC, respectively. In the denitrification column, denitrification started within 2 weeks and total oxidized nitrogen in nitrified leachate (T.O.N between 50 and 150 mgN/l) usually fell below the detection limit at 25°C. No adverse effect was observed on the methanation of waste in the denitrification column with a loading rate of 3.8 gT.O.N-N t-TS_{waste}^{-1} d^{-1}. In conclusion, nitrification in a low-cost biofilter followed by denitrification in a landfill body appears feasible for the removal of nitrogen in landfill leachate in colder climates.

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Acknowledgements

YHTEENVETO (Summary in Finnish)

Kaatopaikan operoinnin ja jätteen esikäsittelyn vaikutus yhdyskuntajätteen biohajoamiseen ja typpipäästöjen hallintaan

Yhdyskuntajätteen käsittely ja loppusijoitus kaatopaikoille on ollut ja tulee olemaan lähivuosina suurten muutosten kohteena johtuen sekä kansallisesta että EU:n jätelainsäädännöstä, joilla edistetään kestävää kehitystä sekä vähennetään jätehuollon aiheuttamia kasvihuonekaasu- ja vesistöpäästöjä. Kaatopaikkojen metaanipäästöjen minimoimisella voidaan selvästi vähentää ihmiskunnan kasvihuonekaasupäästöjä, koska merkittävä osuus päästöistä aiheutuu jätehuollosta ja erityisesti yhdyskuntajätteen kaatopaikkasijoituksesta. Tämän lisäksi suurin osa yhdyskuntajätteen sisältämästä typestä päätyy edelleen kaatopaikkoihin, ja siten esimerkiksi EU:n alueella läjitetään vuosittain noin 2 miljoonaa tonnia jätteen sisältämää typpeä kaatopaikoille. Huolimatta jätehuollossa jo tehdyistä muutoksista kaatopaikkaan varastoituneen typen suotautuminen kestää vähintään sata vuotta.

Yhdyskuntajätteiden kaatopaikoilta muodostuvia kasvihuonekaasu- sekä typpipäästöjä voidaan vähentää sekä lyhyellä että pitkällä aikavälillä yhdyskuntajätteen esikäsittelymenetelmillä ja kaatopaikkajätteen stabiloimiseen tähtäävällä kaatopaikkaoperoinnilla. Esikäsittelyssä tulee kuitenkin huomioida käsiteltävien yhdyskuntajätteiden ominaisuudet sekä esikäsittelyn vaikutukset jätteen ominaisuuksiin. Jätteen biologiseen stabiloitumiseen voidaan päästä, mikäli kaatopaikkaoperoinnissa huomioidaan jätteen koostumuksen lisäksi jätteen biohajoamiseen vaikuttavat biologiset ja fysikaalis-kemialliset tekijät.

Tämän tutkimuksen tavoitteena on ollut selvittää nykyisin käytössä olevien jätteiden esikäsittelyn ja kaatopaikkaoperointimenetelmien vaikutukset kaatopaikoilta aiheutuvien päästöjen minimoimisessa. Tutkitut esikäsittely-menetelmät sisälsivät syntypaikkalajittelun, laitosmaisen mekaanisen erottelun sekä lajittelussa muodostuneiden jätejakeiden ilmastuksen. Kaatopaikkaope-rointimenetelmät sisälsivät jätteen kosteuden vaikutuksen, suotoveden kierrätyksen yhdistettynä pH:n säätöön sekä jätteen esikastelun ennen läjitystä. Kokeet tehtiin laboratoriomittakaavan lysimetreissä. Lisäksi kehitettiin halpaa ja kylmään ilmastoon soveltuvaa suotoveden typen poistomenetelmää, jota voidaan käyttää kaatopaikan suotoveden kierrätyksen yhteydessä.

Erilliskeräyksessä tuotettu kuivajäte sisältää noin 70 % biohajoavia jätejakeita, jotka tuottavat hapettomissa oloissa sekä metanogeenisten mikrobien läsnä ollessa 120 – 320 m^3 metaania (CH_4) kuiva-ainetonnia kohden ja typpeä 0.32-3.5 kg ammoniumtyppeä (NH_4-N) kuiva-ainetonnia kohden. Kuivajätejakeiden ensimmäisen kertaluokan hydrolysaatiovakio vaihteli 0.021 ja 0.058 d^{-1}, jonka perusteella kuivajätteen hydrolysaationopeus on kuudesosa

erilliskerätyn biojätteen vastaavasta. Biojätteen tehokkaalla keräyksellä ja käsittelyllä voidaan selvästi vähentää päästöjä, koska biojäte tuottaa suuria määriä metaania (410 m^3 CH$_4$ kuiva-ainetonnia kohden) ja typpeä (3.6 kg NH$_4$-N kuiva-aine-tonnia kohden). Jätteiden pitkäkestoisella biologisella käsittelyllä esimerkiksi yhdistämällä aerobisen ja anaerobisen käsittely voidaan kaasuntuottopotentiaali saada ennen kaatopaikkaloppusijoitusta alle 20 m^3-biokaasua kuiva-ainetonnia kohden.

Laboratoriokokeissa testatut muuttujat osoittivat, että jätteen hajoamiseen voidaan vaikuttaa kaatopaikkaoperoinnilla. Alhainen kosteus (alle kenttä-kapasiteetin) voi huomattavasti hidastaa, vanhan sekajätteen ja kuivajätteen kohdalla jopa estää jätteen hajoamisen metaaniksi, ja sen vuoksi kontrolloitu kosteuden tai suotoveden lisäys kaatopaikkaan on suositeltavaa, mikäli käytetään vesitiiviitä pinta- ja pohjarakenteita. Kosteuden merkityksen lisäksi kaatopaikkalysimetrikokeet osoittivat, että kaatopaikkaoperointimenetelmät vaikuttivat ratkaisevasti jätteen metanoitumisnopeuteen sekä suotoveden ominaisuuksiin ja siten kaatopaikan operointi bioreaktorina on järkevä menetelmä hajoamisen ja myös päästöjen hallinnassa. Kaatopaikkajätteen nopea hajoaminen metaaniksi nopeuttaa jätteen hydrolysaatiota poistamalla inhiboivat välituotteet ja siten tasapainottaa pH:n neutraaliksi, kun taas jätteen alhainen pH viivästyttää jätteen hydrolysaatiota. Jätteen metanoituminen poistaa jätemassasta orgaanisen aineen (COD) nopeasti, ja siten se lyhentää selvästi kaatopaikan käyttöikää. Nopea kosteuden lisäys kaatopaikka-sijoitettuun sekajätteeseen aiheutti metanaation estävän happokäymisen, joka puolestaan aiheutti korkean ja pitkäkestoisen COD kuormituksen suotoveteen. Suotoveden kierrätys pH:n säädöllä 8:aan oli riittämätön menetelmä jätteen muuttamisessa happamasta metanoivaksi. Kaatopaikalle sijoitettaessa kuivajäte tuottaa huomattavia määriä metaania (70-85 % sekajätteestä) tai hapanta ja väkevää suotovettä mikäli jätettä ei muuteta metaania tuottavaksi esimerkiksi ympin lisäyksellä. Käytetyn neste/kuiva-aine (L/S)-suhteen perusteella voidaan sanoa, että kuivajäte tuottaa suotoveteen korkeita COD ja NH$_4$-N kuormia vähintään 120 vuotta. Hallitulla veden lisäyksellä ja suotoveden kierrätyksellä tämä aika voidaan supistaa muutamiin vuosiin. Metaania tuottavan jätteen ammoniumtypen vapautumista voidaan nopeuttaa lisäämällä kaatopaikan veden virtaamaa siten, että saavutetaan L/S-suhde 3-4. Kaatopaikalle sijoitetun jätteen metaanin tuottopotentiaalia voidaan vähentää myös jätteen hallitulla ilmastuksella, jonka tulee tosin kestää vähintään 51 päivää.

Kaatopaikkojen suotovesien typpi voidaan tehokkaasti poistaa käyttämällä ylöspäinvirtauksella toimivaa nitrifioivaa jätemateriaalista rakennettua bio-suodatinta sekä kierrättämällä nitrifioitu suotovesi edelleen jätepenkereeseen. Murskatusta tiilestä valmistetussa biosuodattimessa 25°C lämpöistä suotovettä voitiin nitrifioida 100-130 mgNH$_4$-N l^{-1} d^{-1} kuormituksella sekä 5°C asteista suo-tovettä 50 mgNH$_4$-N l^{-1} d^{-1} kuormituksella. Kaatopaikkajätteellä täytetyssä de-

nitrifikaatiokolonnissa denitrifikaatio käynnistyi kahdessa viikossa ja siten suotoveden hapettuneiden typen yhdisteiden (TON, yleensä 50 ja 150 mgN l^{-1} välissä) määrä aleni nitrifioidussa suotovedessä 25ºC:n lämpötilassa alle määritysrajan. 3.8 gT.O.N-N/t-TS$_{jäte}$d kuormituksen denitrifikaationopeus ei vaikuttanut jätteen metaanin tuottoon. Siten typen poisto halvalla nitrifioivalla biosuodattimella yhdistettynä nitrifioidun suotoveden denitrifikaatioon kaatopaikkapenkereessä on toimiva menetelmä myös kylmässä ilmastossa.

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