Ammonia Fluxes in Landfills

R&D Technical Report P1-306/TR

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Statement of Use
This output addresses research identified as needed following studies into nitrogen balances in landfills. The research used laboratory studies to formulate a leachate pre-treatment and recycling strategy, and to demonstrate the efficacy of this in reducing leachate ammonia concentrations and accelerating site stabilisation. The report will be of use to waste management practitioners both on the regulatory and operational side, consultants and scientific specialists and is disseminated for information only.

Keywords
Landfill, landfill leachate, leachate recirculation, ammonia, nitrogen transformations, municipal solid waste

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EXECUTIVE SUMMARY

The turnover of the nitrogenous components of landfilled refuse was studied and reviewed in a former Department of the Environment-funded study, 'Nitrogen Balances in Landfills,' available as two publications in the CWM series. In the light of these laboratory studies, suggestions were made of a possible method for managing landfill sites likely to lead to the rapid and permanent reduction of leachate ammonia. This involved a strategy of ammonia treatment and recirculation of nitrified leachate. The aim of this research was to use laboratory studies to formulate a leachate pre-treatment and recycling strategy, and to demonstrate, at the Auchencarroch test cell facility, the efficacy of this in reducing leachate ammonia concentrations and accelerating site stabilisation.

The effectiveness of this strategy was tested in the former DoE-funded landfill test cells at Auchencarroch, Dumbartonshire. A landfill test cell containing pulverised municipal solid waste, designed for enhanced decomposition by leachate recirculation, was fitted with a system that automatically pumped landfill leachate and treated it in an aeration tank before returning it to the landfill. The construction of the aeration tank facilitated the nitrification of ammonia to nitrate which could be removed by denitrification to nitrogen gas within the anaerobic environment of the landfill. Continuous monitoring of gas quantity and quality enabled the effects of the addition of nitrate on methanogenesis to be assessed and the validity of the strategy tested.

Recirculation of treated leachate did not affect gas quality nor did it appear to reduce the gas quantity from before the time that the leachate was recirculated. Methane concentrations and gas flow increased during the study. Ammonia concentrations in the landfill leachate fell during recirculation of treated leachate, and little nitrate could be detected in the test cell during the first year of operation. Continued recirculation without treatment during the winter of 1999 mixed the leachate within the landfill, and ammonia concentrations increased to levels more representative of the start of the experiment. Treatment and recirculation during 2000 led to permanent reductions in landfill leachate ammonia concentrations following routine recirculation without treatment during the winter of 2000 and spring 2001.

Treatment of ammonia was achieved at near optimal conditions without the need for pH control. Overall the procedure treated landfill leachate successfully without compromising gas production. As a strategy the process was viable since it demonstrated that methanogenesis can be maintained during nitrate addition, and the treatment system employed was robust enough to enable effective deployment in the field. Since the study was relatively short in the life of the landfill test cell, some questions still remain about the total nitrogen budget of the test cell and the eventual fate of the recirculated nitrogen, which only a tracer study can confirm.

ACKNOWLEDGEMENTS

Some of the data presented for the test cell analysis comes from routine leachate analyses arranged by EnviroCentre Ltd. and the Caledonian Shanks Centre for Waste Management, Glasgow Caledonian University, and funded by the Department of the Environment and the Environment Agency. We acknowledge the assistance of EnviroCentre Ltd. and the Caledonian Shanks Centre for Waste Management, Glasgow Caledonian University. The authors kindly acknowledge Mr. Ian Wilson and his colleagues at Cunninghame District Council for access to their landfill site at Shewalton. We also acknowledge the kind help of the staff at Glasgow District Council's Summerston Landfill Site.

GLOSSARY

Acetogenesis: The production or formation of acetate in refuse.

Acetogenic phase: The period during the fermentation of refuse when acetate and volatile fatty acids are formed and the pH of the leachate released is usually below 7.

Aerobic decomposition processes: The decomposition of material by microorganisms in the presence of air.

Amonification: The biochemical removal of an amino group (NH_2^-) from protein or other nitrogenous organic compounds.

Anaerobic decomposition processes: The decomposition of material by microorganisms in the absence of air.

Anaerobic bacteria: Bacteria that do not use oxygen for growth and are inhibited by an oxygen rich atmosphere.

BOD (Biochemical Oxygen Demand): A measure of the oxygen used by microorganisms to breakdown organic compounds in a water sample. The higher the BOD the greater the amount of biodegradable compound present in the sample. The BOD is normally measured by comparing the dissolved oxygen concentration in a water sample with that of an identical sample which has been incubated in the dark (to prevent plant growth), at 20^oC for 5 days.

C/N Ratio: The ratio of the carbon content to the nitrogen content.

COD (Chemical oxygen demand): A measure of the amount of oxygen required to oxidise organic compounds present in a water sample, by use of the chemical potassium dichromate. The higher the COD the greater the amount of organic compound in the sample, though a portion of this may be non biodegradable. The method requires the separation of organic compounds from inorganic compounds in the sample which may interfere with the reaction. Measurement of a sample's COD and BOD are rarely equal. However, a sample with a high BOD will typically have a high COD.

Deamination: The biochemical removal of an amino group (NH_2^-) from protein or other nitrogenous organic compounds.

Denitrification: The biochemical reduction of nitrates to nitrogen gas. Can be expanded to include the stepwise reduction of nitrate, nitrite, nitrous oxide and nitric oxide to nitrogen gas during the oxidation of organic compounds by denitrifying bacteria for energy.

Facultatively anaerobic bacteria: Bacteria, which do not require oxygen for growth (but may use it if available), which grow well under aerobic and anaerobic conditions, and to which oxygen is not toxic.

Methanogenesis: The production of methane by anaerobic bacteria.

Nitrification: The biological oxidation of ammonia to nitrite and nitrite to nitrate by ammonia oxidising bacteria and nitrite oxidising bacteria. The process may also be used to describe the release of nitrite and nitrate by certain fungi.

Oxidation: A chemical process involving the combination of oxygen with a compound or substance. The loss of electrons or hydrogen from a substance. Example: Iron (Fe) rusts by combination with oxygen in water to form iron oxide ($Fe_2O_3.H_2O$) also known as ferric III oxide. Iron from its chemical formula has also lost electrons during the oxidation process to become positively charged (Fe to Fe^{3+}).

Reduction: A chemical process involving the removal of oxygen, the addition of hydrogen or the gain of electrons.

1. INTRODUCTION

Current or 'dry tomb' landfilling practices contradict many of the principles of sustainable development. Not only are valuable resources lost through consumption and landfilling but the waste will still being hazardous to the environment for decades. At the time of writing, waste minimisation and recycling will be the primary objective of the UK Government as the EC Landfill Directive (1999/31EC) is implemented. However, landfill will still be the primary disposal route for the majority of UK waste at least in the foreseeable future or until suitable alternatives have been found. One alternative is to encourage sustainable landfill technology, which offers both the capacity of current landfilling practices but could also meet environmental concerns by rapidly attenuating the waste. The enhanced decomposition rates also offered significant renewable energy production in the form of landfill gas.

To meet the criteria demanded by sustainable development, landfilling must meet the needs of the present without compromising the ability of future generations to meet their own needs (Bruntland Report, 1987). Sustainable landfilling strategies such as the 'Landfill Bioreactor' (EPA, 1995) or the 'Flushing Bioreactor' (Harris *et al.*, 1994) concepts extol rapid refuse decomposition, coupled, in the latter, with a flushing action to remove unwanted toxic pollutants. Of principal importance to the success of these landfill strategies is a moist, homogenised and uniform refuse, enclosed within a system which allows for efficient water distribution. Moist refuse degrades rapidly in comparison to dry refuse allowing for enhanced bacterial activity with subsequent increases in the rate of gas production and refuse stabilisation.

The accelerated decomposition should allow the refuse to stabilise to a point that it is does not pose a risk to subsequent generations after 30 years. The landfill gas produced during decomposition must be used as a power generation resource so that the waste is reused. Contradictions with sustainable development lie with the landfilling of materials that ultimately end up in the waste. These can, or should be, when beneficial to the environment, be recycled and reused. Further contradictions are associated with the large amount of land required for landfilling, land that might potentially be used for other purposes.

1.1 Enhanced landfill gas production

Landfill gas is a renewable energy source. The enhancement of gas production by landfills was first explored in the United Kingdom through a number of test cell experiments and laboratory studies, funded by local councils, the Department of the Environment, and latterly the Environment Agency. Guidelines for accelerated stabilisation were issued in Waste Management Paper 26B. This discusses the features of a site designed for enhanced stabilisation, based on current knowledge about the decomposition of municipal solid waste (MSW). The major research projects are listed below:

1.1.1 The managed landfill research programme

This Department of the Environment funded project was carried out between 1978 and 1983 and explored the effect of different waste densities following compaction, recirculation of leachate and water, daily cover and capping materials and co-disposal of hazardous wastes. A total of eight test

cells were constructed, each containing approximately 3,500 tonnes of waste to a depth of 5m. Dry pulverised domestic waste was also landfilled (Campbell, 1985).

1.1.2 Landfill 2000

The Landfill 2000 experiment (WRc, 1995) was initiated by West Yorkshire Waste Management and Yorkshire Water Enterprises. The aims of the experiment were to establish optimum conditions that would give a three year waste degradation period, quantify the methane generated and determine the biological condition of the waste and its use as an environmentally friendly soil conditioning medium. Additional support was provided by the Department of the Environment through the Education Training Support Unit (ETSU), appointing WRc plc with the aim of carrying out the process monitoring requirements for the project.

Two similar test cells, 36x23m with a maximum central depth of 5m, were constructed in 1991. Cell 1 contained 930 tonnes domestic refuse and 135 tonnes sewage sludge cake and cell 2, used as a control, contained 980 tonnes domestic refuse and 127.5 tonnes sewage sludge cake. The cells were packed to density of 0.94 and 0.89 tonnes m^3 respectively. Cell 1 was operated with leachate recirculation, and the gas production and leachate quality compared with cell 2.

1.1.3 The Brogborough test cells

The investigation of landfill as an energy resource began in earnest in 1986 when the Brogborough landfill test cells were designed and constructed. The six test cells were built with the aim of determining the factors most likely to enhance landfill gas production. The cells measured 25x40x20m when filling was finally completed. The cells were capped in the spring of 1989. The six cells consisted of a municipal solid waste (MSW) control, low density MSW, MSW following water addition, MSW with air injection, 93% MSW with 7% sewage sludge and 55% MSW with 45% non hazardous industrial waste.

Gas production has been measured from the cells since 1989. The cells were constructed by AEA Technology with funding from the Department of Trade and Industry. One weakness of the Brogborough experiment is the lack of pretreated or pulverised refuse in their construction, which could hinder the decomposition of the refuse. Otherwise, owing to their depth, they represented an extremely useful model landfill experiment. A review of the data up to 1994 was produced by Knox (1995a). The project finished in 2000.

1.1.4 Field trials of waste manipulation techniques (The Auchencarroch Test Cells)

Four test cells (30 x 28 x 5m) were constructed for the former Department of the Environment with the aim of determining the effect of waste pre-treatment on landfill gas production. They each contain approximately 3800 tonnes of MSW, covered with a 1.2m clay cap and enclosed under 0.9m soil cover. The cells contain either a mixture of pulverised refuse and sand (27% by volume), pulverised refuse or unpulverised refuse (EnviroCentre Ltd., 1998). A further test cell containing unpulverised refuse acted as a control and was the only test cell in which leachate was not recirculated.

The principal weakness of the cells was the lack of a pulverised unrecirculated control, and their depth, which was limited to 5m. They were also constructed in a cold mountainous area. They were originally constructed and run by the Civil Engineering Department of the University of Strathclyde, its commercial arm (The Centre for Environmental Management Studies now EnviroCentre Ltd.) and a local landfill operator (George Munn). In 1998 a new contract to maintain the test cells was awarded to Glasgow Caledonian University. This contract will end in 2001.

1.1.5 Predicted landfill gas yields from municipal solid waste

Based on an American study the reported gas production from refuse varied from 120 to 410 m^3 $tonne^{-1}$ dry waste (Pohland & Harper, 1986). In 1995 the most productive MSW Brogborough test cells were producing gas at a rate of 18-22 m^3 t^{-1} emplaced waste $anum^{-1}$ (Knox, 1995a). Short term laboratory studies with 1-2 month old landfilled pulverised MSW produced 2.5l gas from 53.2g dry weight equivalent to 47m^3 $tonne^{-1}$ dry weight. Total methane production was equivalent to 28.2 m^3 $tonne^{-1}$ (Burton & Watson-Craik, 1997). Wolffson (1985) produced figures of 216, 291, 94, 81, 250 and 24 m^3 gas $tonne^{-1}$ dry weight for grass, vegetables, newspapers, magazines, cardboard and sawdust respectively (Stegmann & Spendlin, 1987). Approximately 200 m^3 $tonne^{-1}$ fresh waste is a rough guideline figure for MSW in a landfill bioreactor.

1.2 Sustainable landfilling in the context of the EC Landfill Directive

There is much evidence in support of the acceleration of the landfill decomposition processes by the use of wet landfill bioreactors (Reinhart & Townsend, 1998). The waste within large dry tomb landfills shows little decomposition and can, for example, be easily dated from buried newsprint since little actual decomposition has taken place. Landfill gas can be utilised for power generation. In June 2000 the UK had a current operational capacity of 302 MW, enough to supply 350,000 households or a city the size of Leeds (Biogas, 2000). It remains to be seen whether these arguments for landfilling and sustainable landfilling can be renegotiated within the context of the recently ratified EC Landfill Directive (1999/31/EC).

The directive requires a reduction in the biodegradable waste sent to landfill of 75% of total biodegradable municipal waste (weight) produced in 1995 by 2006, a reduction to 50% by 2009 and a reduction to 35% by 2016. The directive does not fix the way in which these reductions have to be obtained. However, it gives a clear indication that biodegradable waste should be preferably treated via biological treatment processes such as composting and anaerobic digestion. The EC Landfill Directive (1999/31/EC) will also bring to an end the practice of co-disposal (where hazardous and non-hazardous wastes are disposed of in the same landfill).

The landfilling of hazardous materials will only be possible in facilities specifically designed to accept the materials. There will be an increased requirement for pre-treatment of waste prior to landfilling, a banning of certain materials completely from disposal to landfill, if they possess for example, corrosive, oxidising, flammable, or liquid properties (DETR, 2000). The special wastes would have a greater capacity to pollute, which would in turn inhibit their own breakdown, unless they were artificially stabilised by encasement. However, planning constraints are likely to limit the rapid uptake of alternative waste disposal methods to landfilling in the short term.

Discussion documents on the Biological Treatment of Biodegradable Waste from the European Commission, circulated for consultation on 20th October 2000 (DG ENV.E.3/LM/biowaste/1 st draft) make no mention of landfill bioreactors and only concern themselves with anaerobic digestion and composting as methods for biologically degrading degradable waste. What may be required of an anaerobic digestion plant or a possible future landfill bioreactor/anaerobic digestion plant hybrid is detailed in the following section (see 1.2.1). Of the bioreactor experiments carried out in the UK only Landfill 2000 (see 1.1.2) was constructed with the bold stated aim of producing a compost like end product following the anaerobic fermentation of the waste.

1.2.1 Consultation Document on the Biological Treatment of Biodegradable Waste (DG ENV.E.3/LM/biowaste/1st draft)

'The anaerobic digestion process shall aim at the anaerobic decomposition of biodegradable waste under controlled conditions, its reconstitution into humus by the action of micro-organisms (methanogenic bacteria) as well as the production of biogas. The anaerobic digestion treatment of biodegradable waste should have the purpose of reducing the fermentability of this waste, maximise the production of biogas, and ensuring that the digestate can be used for the production of compost. It should be carried out in such a way as to minimise the impact on the environment of air emissions and leaching to surface or groundwater as well as to minimise the health impact for aerosols on the workers at the plant.

The liquor from an anaerobic digestion plant shall be suitably treated to comply with the requirements of Directive 91/271/EC before being released into surface water. The anaerobic digestion process shall be carried out in such a way that a minimum temperature of 55 °C is maintained over a period of 24 hours without interruption and that the hydraulic dwell time in the reactor is at least 20 days. In case of lower operating temperatures or shorter period of exposure, the biodegradable waste shall be pre-treated at 70 °C for 1 hour, or the digestate shall be post-treated at 70 °C for 1 hour, or the digestate shall be composted.'

1.3 Sustainable landfilling through combined nitrification and denitrification

The acceleration of the decomposition processes can be achieved by increasing the moisture content of the waste. It can also be achieved by increasing the moisture availability through the recirculation of leachate collecting in the sump. This has been shown to significantly accelerate refuse decomposition (Robinson *et al.*, 1982, Tittlebaum 1982, Barlaz *et al.*, 1987). Leachate recirculation offers a reduction in the chemical oxygen demand (COD) (see glossary) and biological oxygen demand (BOD) (see glossary) through enhancement of methanogenesis (see glossary), although residual COD persists and requires dilution or treatment prior to discharge.

Whilst recirculation of landfill leachate can facilitate greater moisture availability it can initially lead to the rapid solubilisation of toxic waste products from the decomposition processes (Appendix 1), and the creation of a leachate containing high concentrations of volatile fatty acids and ammonia. These ammonia concentrations may persist long into the life of the landfill and may, in combination with high initial volatile fatty acid levels, reduce the benefits of using recirculation to accelerate

decomposition. There appears to be no major sink for ammonia in landfill leachate since the anaerobic decomposition processes (see glossary) do not assimilate nitrogen into biomass to the same extent as aerobic processes (see glossary). Anaerobic decomposition within a landfill will lead to modest decreases in the biodegradable portion of waste through methane and carbon dioxide production, ammonia, and other less toxic mineral ions persist in the leachate (Robinson, 1995).

Despite the relatively high carbon to nitrogen ratios in municipal solid waste, ammonia is released through protein degradation (Burton & Watson-Craik, 1996ab). The ammonia requirements for bacterial growth during anaerobic decomposition of refuse are modest, and the excess nitrogen from protein decomposition is released into the leachate (Burton & Watson-Craik, 1998). Ammonia concentrations of approximately 3000 mg $NH_{3/4+}$-N l^{-1} can significantly inhibit methanogenesis (Burton & Watson-Craik, 1998). Leachate ammonia concentrations in landfill leachate decrease extremely slowly following closure (Kruempelbeck & Ehrig, 1999). Pumping and treating leachate aerobically in lagoons may represent the main route for rapidly removing toxic ammonia from landfill sites.

If the toxic products could be treated aerobically during recirculation then this may alleviate the inhibition of the anaerobic decomposition processes, and in particular the inhibition of methanogenesis (Burton and Watson-Craik, 1998). The process may offer a complete treatment strategy for ammonia through combined nitrification and denitrification (see glossary) (Figure 1.1), whilst maintaining the moisture content of the landfill. Used in tandem with current recirculating bioreactor thinking (EPA, 1995; Harris *et al.*, 1994, Knox, 1995b), it could offer both a flexible approach to leachate management, and reduce ammonia in the landfill by treatment and dilution (Figure 1.1). Overall, the strategy strives to promote the overall degradation of the waste and landfill gas and energy production.

Nitrification is a biologically mediated reaction performed on the most part by ammonia oxidising bacteria and nitrite oxidising bacteria. It is used during aerobic treatment of wastewaters to remove toxic ammonia and convert it into nitrate which is less toxic. The bacteria oxidise either ammonia or nitrite, and excrete nitrite (as nitrous acid) or nitrate respectively. The process is acidic. Denitrification is an anaerobic process performed by anaerobic bacteria in which nitrate or other oxidised forms of nitrogen such as nitrite, nitric oxide and nitrous acid are converted or reduced (see glossary) to nitrogen gas. The process consumes organic materials which are oxidised (see glossary) into smaller compounds. The process is alkaline. Together nitrification and denitrification can be employed in sequence to treat ammonia to nitrogen gas in wastewaters. Separate vessels containing oxygen rich and oxygen free wastewaters are required. The denitification process requires a suitable oxidant, and in tertiary anaerobic treatment, methanol is often supplied to promote denitrification of nitrate in leachates.

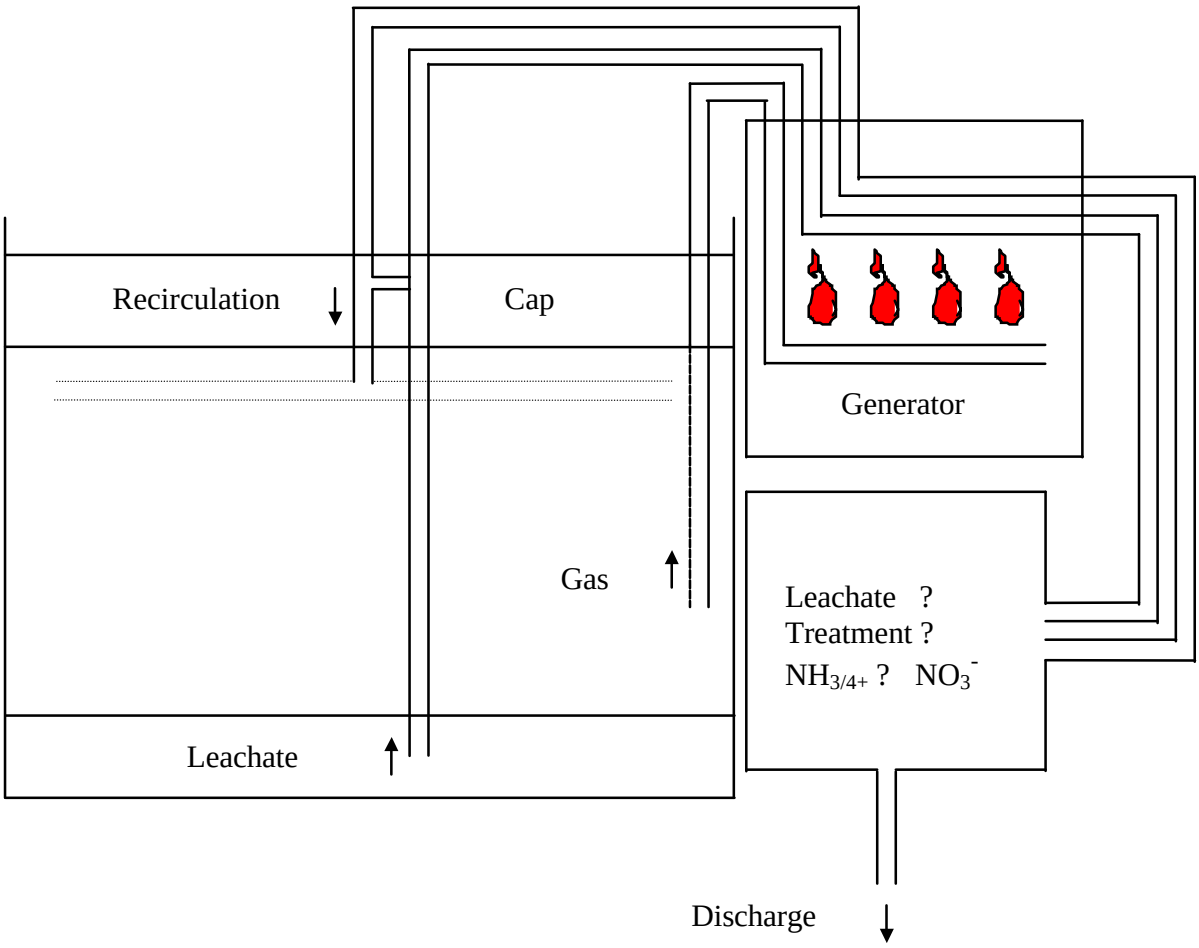


Figure 1.1 Schematic diagram of the wet/flushing bioreactor with combined leachate treatment recirculation

Previous work in the laboratory indicated that landfilled refuse readily denitrified. Pulverised refuse cultures could be induced to produce nitrous oxide from nitrate by a headspace of 10% acetylene gas (Sinclair, 1994), that block the reduction of nitrous oxide to nitrogen during the bacterial process of denitrification (Yoshinari & Knowles, 1976). Autoclaving stopped nitrous oxide production, identifying a biological process. The addition of nitrate to refuse cultures demonstrated the ability of landfilled refuse at 80% (w/v) moisture content to denitrify. Nitrate was lost from the systems containing 500 and 1000 mg NO_3^--N l^{-1} within 6 days without significant increases in ammonia concentration (Burton & Watson-Craik, 1996a). It was concluded that the vast majority of the nitrate was reduced to nitrogen gas (N_2) or nitrous oxide (N_2O). Denitrifying bacteria were present in refuse in sufficient numbers to initiate rapid denitrification and nitrate consumption despite the complete absence of nitrate in 1-2 month old landfilled refuse (Burton & Watson-Craik, 1997). Nitrate can be identified in vegetable and organic matter and soils but often the bulk concentrations are too low to be detected in refuse extracts. Whilst nitrate in the refuse may be reduced still further as the refuse ages, denitrifying bacterial species are maintained and will proliferate following the addition of nitrate.

Nitrate concentrations of 500 and 1000 mg NO_3^--N l^{-1} inhibited methanogenesis by refuse cultures at 80% and 95% moisture content despite low initial redox potentials (-200 mV or less). Only after six days when the nitrate was exhausted did methanogenesis recover. In previously methanogenic cultures approximately 20 days were required before increases in methane production could be determined. Lower concentrations of 50 mg NO_3^--N l^{-1} stopped methane production for approximately 20h, and methanogenesis completely recovered after 3 days incubation. However, at these lower concentrations some 4-7% of the nitrate was reduced to ammonia. Methanogenesis by bacterial cultures enriched from the refuse proved resistant to concentrations of 5.7 mg NO_3^--N l^{-1} and both methanogenesis and denitrification could proceed together in liquid culture (Burton & Watson-Craik, 1996b).

In lysimeter experiments performed by Knox (1995b) refuse of various ages was tested for its ability to denitrify treated and nitrate containing leachate. Young and old refuse of an initial fresh weigh of 1680 and 2330kg at densities of 772 and 1074 kg m^{-3} respectively were supplemented with leachates containing nitrate. Young refuse was capable of denitrifying nitrate completely at a rate of 15g NO_3^--N m^3 day^{-1} for approximately 6 bed volumes. Recirculating leachate to initiate methanogenesis enabled methanogenic conditions to develop. Denitrification could continue in the presence of methanogenesis during further leachate purturbations containing nitrate concentration of 500 mg NO_3^--N l^{-1}. Ammonia concentrations in the effluent were not believed to increase as a result of nitrate consumption by the refuse. Older refuse, between 40-70 years old, failed to support denitrification.

Nitrification of ammona and leachate COD reduction could be achieved in a lagoon prior to recirculation. Such a strategy would enable the nitrification of ammonia ex situ (Knox, 1985) and the denitrification of nitrate in the landfill (Knox & Gronow, 1995) whilst enhanced decomposition of recalcitrant organic compounds by aerobic decomposition could occur in the lagoon. The strategy would promote the rapid decomposition of landfilled refuse through recirculation and moisture control, and offer a complete treatment strategy for ammonia for both present and future landfill practices. The system would be dependent on combined nitrification and denitrification for ammonia treatment.

The effect of recirculating nitrified leachate may have serious consequences for methanogenesis and this was investigated in field scale experiments at the Auchencarroch Landfill Site, Dumbartonshire. Using a test cell constructed for recirculation studies on pretreated waste, a batch fed biologically aerated filter was constructed to treat landfill leachate. The treated leachate was returned to the landfill and the gas production compared to previous data held for the site.

1.4 Aims

Before a strategy of combined leachate treatment and recirculation can be attempted, the effects, on the decomposition processes, of nitrified leachate (containing nitrate) must be elucidated. The objective of this programme was to provide information on the effects of the addition of nitrate to a landfill site following leachate treatment. The data collected was used to determine the viability of combined leachate nitrification and landfill denitrification as a method for the acceleration of landfill decomposition and ammonia treatment. The performance of the constructed aeration tank used to treat landfill leachate was measured and the effects on landfill gas production monitored. The ability of the refuse to denitrify was demonstrated. The experiment complements recently funded work by the former Department of the Environment and the Environment Agency on nitrogen balances in refuse, denitrification and contaminant flushing and the potential pollution load from refuse.

2. GENERAL MATERIALS AND METHODS

2.1 On site analysis

Routine assessment of leachate nitrate and ammonia concentrations were performed using Merkoquant test strips and Reflectoquant test strips and their associated reader (Merk Eurolab). Leachate pH was determined using a MTW data logging pH probe. Dissolved oxygen concentrations were measured using a Jenway dissolved oxygen electrode. Leachate levels were measured using a Geotechnical Instruments meter. Analysis of the relative concentrations of methane, carbon dioxide, oxygen and hydrogen gas were performed using a Geotechnical Instruments GA-90 infrared gas analyser. Liquid samples were taken during routine recirculation or following pumping of leachate. To determine whether nitrate or rainwater water was entering the central well, samples were removed manually using Waterra tubing.

2.2 Analysis of landfill leachate by Bodycote Ltd.

The routine analysis of landfill leachate from Auchencarroch Test Cell 3 was performed by Bodycote Ltd., Coatbridge, Glasgow. The leachate was collected from the sump of the test cell during recirculation or by extraction using Waterra tubing. The use of this service has been continuous since 2.12.98. From 1999 it was organised by the Caledonian Shanks Centre for Waste Management, Glasgow Caledonian University, who held the Environment Agency contract for the maintenance the Auchencarroch Test Cells. The analysis was performed at the same time as the other three Auchencarroch test cells, and continued the original Field Trials of Waste Manipulation Techniques project (EnviroCentre Ltd., 1998). The data were collated by the authors and the Caledonian Shanks Centre for Waste Management. Analysis data pertaining to the Auchencarroch Test Cell Project will be submitted in a forthcoming report by the Caledonian Shanks Centre for Waste Management and published by the Environment Agency. The report will also include analysis data after 2000 collected by the Caledonian Shanks Centre for Waste Management.

2.3 Laboratory analysis

Analysis pertaining to the Ammonia Fluxes in Landfills project was carried out at the University of Strathclyde.

2.3.1 Clarification of the refuse sample

Where spectrophotometric analysis was required, the clarification of the liquid sample was carried out prior to performing the assay. A refuse sample (20 g) was adjusted to the appropriate moisture content and volume (70-100 ml) with distilled water and mixed by blending in a liquidiser for 5 min or by shaking in a stoppered flask for 30 or 60 min held on a Griffin flask shaker. The bulk solids were separated by passage through a 5.5 cm Whatman GF/C glass micro-fibre filter (1.2 mm retention) held on a Buchner funnel under vacuum. Dirty suspensions and culture samples containing sulphide were filtered twice. Bacterial cells were removed by centrifugation at 8000x gravity.

2.3.2 Analysis of Nitrogenous Compounds in Refuse

All chemicals were obtained from BDH Merck Ltd unless stated otherwise.

2.3.3 Spectrophotometric analysis of nitrite

Nitrite ions react with sulphanilamide and N-1-naphthylethylenediamine in orthophosphoric acid to form a red azo-dye whose concentration can be determined spectrophotometrically at 540N.M.

Macro-determination of nitrite

Clarified samples, not exceeding 40 ml, were decanted into 50 ml volumetric flasks. To each flask 1 ml of sulphanilamide reagent was added, mixed and diluted to 50 ml. The solution was incubated at 25 ^{o}C for 1 h and the absorbance measured at 540 N.M. and compared to standard aqueous solutions of nitrite (0 - 50 mg NO_2^--N l^{-1}).

The sulphanilamide reagent was prepared by dissolving sulphanilamide (40 g) in a mixture of 100 ml of orthophosphoric acid (85% m/v) and 500 ml distilled water. N-1-napthylethylenediamine dihydrochloride (2.0 g) was then dissolved in the solution which was then made up to 1 l and stored at 4 ^{o}C in an amber bottle.

Micro-determination of nitrite

To 1 ml of a clarified sample and 1ml distilled water in a 1 cm cuvette was added 40 µl of sulphanilamide reagent. The reagent was prepared by dissolving sulphanilamide (5 g) in a mixture of 12.5 ml of orthophosphoric acid (85% m/v) and 125 ml distilled water. N-1-napthylethylenediamine dihydrochloride (0.25 g) was dissolved in the solution which was then made up to 250 ml and stored at 4 ^{o}C in an amber bottle. The solution was incubated at 25 ^{o}C for 1 h and the absorbance measured at 540 N.M. and compared to standard aqueous solutions of nitrite (0 - 3 mg NO_2^--N l^{-1}).

2.3.4 Spectrophotometric analysis of ammonia

Ammonia concentrations were determined colorimetrically. Stock reagents of phenol: alcohol (10 g phenol in 100 ml of 95% methanol), nitroprusside (0.5 g of sodium nitroprusside in 100 ml of distilled water), alkaline citrate (100 g of sodium citrate plus 5 g of sodium hydroxide in 500 ml of distilled water) and 15% w/v sodium hypochlorite solution were prepared, stored at 4^{o}C and used within 1 month. To 1 ml of a clarifed sample in a plastic cuvette was added, 1 ml distilled water, 40 µl of phenol solution, 40 µl of nitroprusside solution and 100 µl of oxidising solution (prepared by adding 10 ml of alkaline citrate to 2.5 ml of hypochlorite solution). The reaction mixture was incubated at 50 ^{o}C for at least 1 h at 50^{o}C or left overnight before reading the absorbance at 640 nm. A calibration curve was constructed with 0 - 3.0 mg $NH_{3/4+}$-N l^{-1}. Using the same reagents, the assay was adapted for use with 4ml capacity cuvettes in which case 1.5 ml of sample, 1.5 ml of water, 60 µl of phenol reagent, 60 µl of nitroprusside and 150 µl of oxidising solution were used.

2.3.5 Spectrophotometric analysis of ammonia using Nessler's Reagent

The spectrophotometric analysis of ammonia was carried when there were high concentrations of ammonia present but limited concentrations of interfering mineral ions in solution. To a clarified sample (2 ml) in a cuvette was added 30 µl of Nessler's Reagent. The mixture was stirred, left for 20 min and the absorbance measured at 425 nm using a spectrophotometer and compared to a calibration curve (0 - 4.0 mg $NH_{3/4+}$-N l^{-1}).

2.3.6 Analysis of total nitrogen content by the Kjeldahl method

Refuse samples (20 g) of known moisture content were adjusted to the appropriate moisture content and volume (70 - 100 ml) with distilled water and mixed by blending in a liquidiser for 5 min. Suspensions equivalent to 2 g refuse (dry weight) were placed in macro-Kjeldahl glass digestion tubes containing 3.67 g Kjeldahl catalyst (mixed/powdered 10 g K_2SO_4, 1 g $CuSO_4.5H_2O$ and 0.1 g Se). Concentrated sulphuric acid (10 ml) was added and the suspension was digested for 60 min on a Tecator 2020 thermostatically controlled heating block with fume extraction according to the manufacturer's instructions.

The digested samples were allowed to cool to room temperature, diluted with 50 ml distilled water and distilled on a Tecator 1026 distilling unit after automatic neutralisation with 50 ml 40% (w/v) NaOH. The distillate was collected in a 250 ml Erlenmeyer flask containing 25 ml boric acid solution (40 g boric acid, 10ml bromocresol green, 0.1% w/v in ethanol), 7ml methyl red (0.1% w/v in ethanol and diluted to 1 l with distilled water). The collected distillate was titrated with 0.1 M HCl and the percentage nitrogen present in the sample calculated from the following equation as documented in the manufacturer's instructions.

$$\%N = \frac{14.007 \times (\text{ml HCl titrant} - \text{ml HCl titrant blank}) \times \text{molarity of standard acid}}{\text{sample mass} \times 10}$$

The reagent blank was measured using 2 g sucrose of low nitrogen content instead of refuse samples.

2.3.7 Qualitative analysis of nitrogenous compounds in refuse

Qualitative analysis of nitrate (10-500 mg NO_3^- l^{-1}) was carried out using Merckoquant test sticks (BDH Merck). In addition, qualitative analysis of nitrite (1 mg NO_2^--N l^{-1} and above) could be achieved by adding one drop of Greiss-Ilosvay's reagents 1 and 2 to 0.5 ml of liquid sample. A red colour signalled the presence of nitrite. The method could be modified to measure nitrate in the absence of nitrite by placing a few grains of Zinc powder to the sample for 5 minutes to reduce some of the nitrate to nitrite before addition of the Greiss-Ilosvay's reagents. Ammonia could be detected by adding a drop of Nessler's reagent to 0.5 ml of the liquid sample, a positive result was indicated by an orange colour.

2.3.8 Sulphate analysis (Rand et al., 1975)

Sulphate analysis was achieved using the turbidometric method of Rand *et al.* (1975). A clarified sample (10 ml) was decanted into a screw top bottle and oxygen free nitrogen bubbled to expel any sulphides. To this was added 0.5 ml of conditioning reagent (glycerol, 50 ml; concentrated HCl, 30 ml; 95% v/v isopropyl alcohol, 100 ml; NaCl, 75 g; glass-distilled water, 300 ml). A standard spoonful (approximately 0.6 g) of $BaCl_2$ crystals, dry, 20-30 mesh) was added and the resulting solution mixed immediately on a vortex mixer for 60 s. After 4 minutes the solution was poured into a cuvette and the absorbance of the $BaSO_4$ precipitate read at 420 nm with a spectrophotometer.

Sodium sulphate was used as the standard, with a concentration range between 0.1 and 1 mM. A standard curve was derived on each occasion from which the sulphate concentrations of the samples were determined. A reduced volume method was also used successfully in 1.5 ml Eppendorf centrifuge tubes, reducing the sample volume required for each analysis.

Correction for sample coloration and turbidity was achieved by running blanks to which the $BaCl_2$ was not added.

2.3.9 Measurement of conductivity

Conductivity was measured using a Hanna Conmet and a Jenway 4320 conductivity meter.

2.3.10 Measurement of volatile fatty acids/carboxylic acids and methane

Samples (0.9 ml) of leachate were acidified with 0.1 ml formic acid (Aristar Range, BDH). One µl was injected into a Perkin Elmer 8500 Gas Chromatograph linked to a personal computer with a 900 series Perkin Elmer Interface, and containing a 2 m glass column (internal diameter 2 mm) containing 5% neopentyl glycol sebacate + 1% H_3PO_4 on Chromosorb W - AW (80-100 mesh) (Phase Separations Ltd, Clwyd). The injector and detector temperatures were set at 210^o and 220^oC respectively. The oven temperature was set for 2 minutes at 125^oC with a ramp rate set at $25^oC\ min^{-1}$ to reach a final oven temperature of 160^oC where it was held for 1 minute. Peak areas were compared to a similarly acidified standard of 10 mM solution of acetate, propionate, iso-butyrate, butyrate, iso-valerate, valerate and hexanoate. Linearity of peak area against concentration was confirmed across the concentrations measured. Analysis was confined to these seven volatile fatty acids.

Methane analysis was achieved using the same operating procedure for volatile fatty acids except gaseous sample injections of 50 µl were made and compared with pure methane injections (BOC gases) of 10 µl. The injector, detector and oven temperatures were set at 200^o, 210^o and 80^oC respectively.

2.3.11 Measurement of chemical oxygen demand (COD)

Measurement of Chemical Oxygen Demand was performed on clarified leachate samples using a Hach COD reactor and Hach DR2010 spectrophotometer system. Low range (0-1000 mg O_2 ml^{-1}) and high range (0-10,000 mg O_2 ml^{-1}) commercially prepared COD tubes were used (Camlab, Cambridge).

2.3.12 Analysis of leachate by ion chromatography

Recent samples (2001) of leachate from the constructed aeration tank were measured in 2001 using a dual column Dionex DX-120 ion chromatograph with AS-14 and CS-12A columns. Clarified and filtered leachate was diluted by 50 or 100 fold and 25µl injected by autosampler at a flow rate 1.1 ml min^{-1}. Concentrations of lithium, ammonia, sodium, magnesium, potassium, calcium, ammonia, nitrate, nitrite, chloride and sulphate were determined.

3. LABORATORY STUDIES ON LEACHATE TREATMENT

Laboratory studies were undertaken to test the requirements of the treatment of leachate from the landfill test cell chosen for the recirculation study. The laboratory systems were designed to reproduce the predicted operation of the Auchencarroch aeration tank. Leachate, 100 ml, from the Glasgow District Council Summerston landfill leachate treatment lagoon (10% solids) was mixed with 300ml leachate from the landfill test cell in a 1l glass bottle (Duran) and incubated in an insulated water bath maintained at $15^{o}C \pm 1^{o}C$ by a Churchill thermo-circulator. Aeration (1 litre h^{-1}) was achieved using a Hy-Flow air pump connected to a 5mm internal diameter stainless steel aeration tube. All tubing and connectors were either silicone rubber or polypropylene.

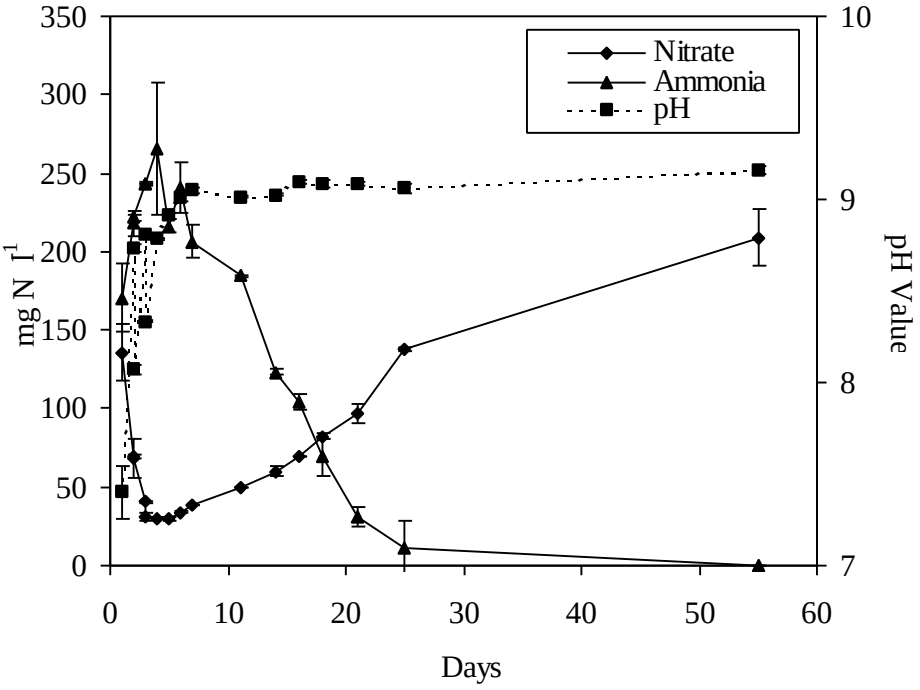
After 3 and 6 days a further 300ml of leachate from the test cell was dispensed to a final volume of 1l to simulate the pulsed addition of leachate. The leachate was then aerated for a further 7 weeks. Samples (5 ml) were removed for analysis. Reflectoquant test strips and meter (Merck) were used to analyse ammonia and nitrate. Concentrations of ammonia were also measured spectrophotometrically by reaction to indophenol blue with bleach and phenol. Nitrite concentrations were analysed spectrophotometrically by reaction with sulphanilamide and N-1-napthylethylenediamine (Burton and Watson-Craik, 1997).

3.1 Results of the laboratory studies on leachate treatment

Nitrification was recorded in laboratory scale studies at $15^{o}C$ on leachate from the landfill test cell. Nitrate concentrations increased during aeration (Figure 3.1). Discrepancies between initial ammonia concentration and the nitrate concentration after 25 days could be explained by accumulation of nitrogen in the biomass since allowing the leachate to continue aerating for a further 25 days caused the bacterial biomass to sink to the bottom, and to degrade and liberate ammonia for further nitrification. Other small discrepancies may be accounted for by ammonia volatilisation, biomass accumulation or other microbial processes. There was no significant accumulation of nitrite in the cultures.

It was considered that there might be implications for the successful enhancement of the decomposition processes in the landfill should additional supplements be required to adjust the pH value or phosphate concentration during treatment. Any additions would contribute to the ionic strength of the leachate and eventually the landfill. Laboratory experiments confirmed that nitrification could be facilitated without any additional supplements, and more importantly the aeration of the leachate promoted an increase in pH value to buffer the nitrification process. Volatile fatty acids (C_2-C_6) could not be detected in the leachate prior to aeration indicating that the recirculation performed during the operation of the landfill test cells had succeeded in removing the organic components normally observed during the acetogenic phase of landfill decomposition.

Figure 3.1 Nitrification of landfill leachate from Auchencarroch Cell 3 seeded with treated landfill leachate from a treatment lagoon and incubated at 15°C. Error bars represent the standard deviation of duplicate cultures.



4. RECIRCULATION OF NITRIFIED LEACHATE THROUGH A LANDFILL TEST CELL

4.1 The Auchencarroch Test Cell Experiment

4.1.1 Introduction

For a complete description of the Auchencarroch Test Cell Experiment the reader is referred to the original contractor report submitted to the Department of Environment by EnviroCentre Ltd., Field Trials of Waste Manipulation Techniques (CWM 173/98). A paper was also presented at the 6th International Landfill Symposium, 13-17th October, St. Margherita di Pula, Cagliari, Sardinia. (Wingfield-Hayes *et al.*, 1997).

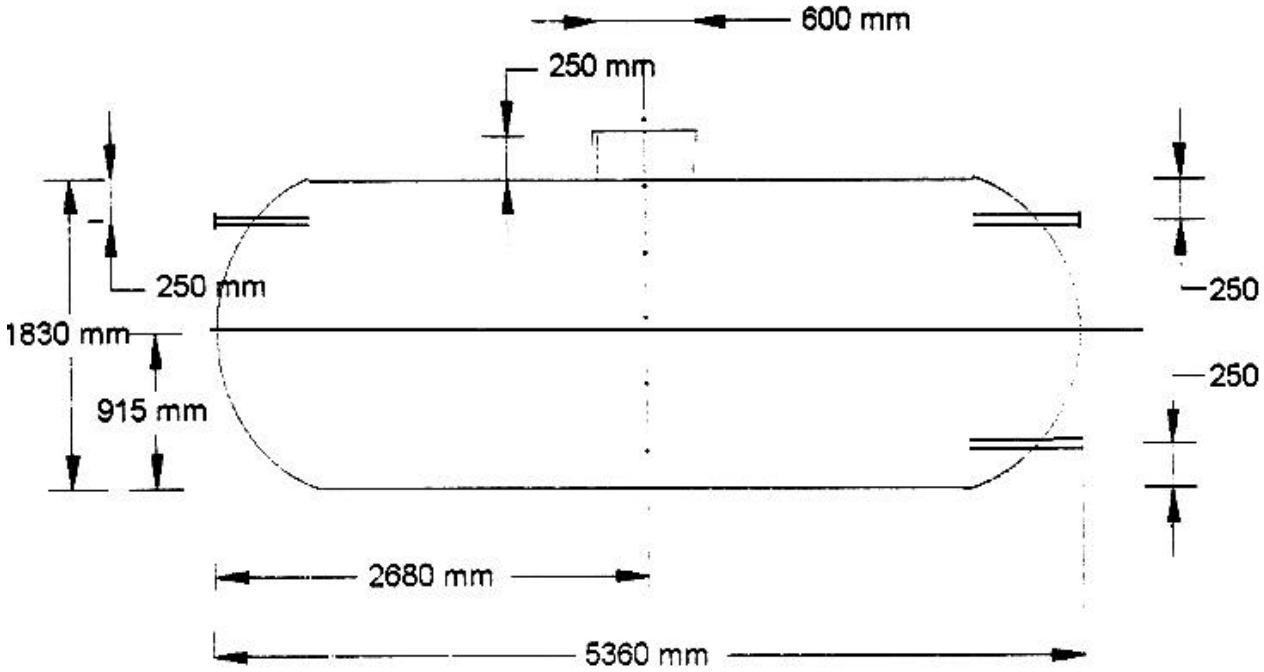
The test cell (Auchencarroch Test Cell 3) chosen for the study (30 x 28 x 5m) contained approximately 3800 tonnes of pulverised MSW diverted from the Cunninghame District Council landfill site at Shewalton. Pulverisation was achieved by the use of a Dano drum in which refuse was shredded in a rotating drum. The refuse was covered with a 1.2m clay cap and enclosed under 0.9m soil cover. Following the end of the DoE funded project Field Trials of Waste Manipulation Techniques (CWM 173/98) in October 1997, the chosen test cell was recirculated each month for 1 h at a rate of 0.125 m^3 min^{-1} (total volume 7-7.5m^3). Leachate was recirculated from the central well into a granular recirculation blanket of crushed rock (150mm deep), separated from the cap above by a geotextile membrane.

A gas collection pipeline on the surface of the refuse draws gas through a Bartington Associates Ltd. gas flow meter (EnviroCentre Ltd., 1998). The flow meter is an updated version of the flow meter developed during the Waste 2000 project (WRc, 1995). The gas was analysed monthly for carbon dioxide, methane and oxygen using a Geotechnical Instruments GA90 infrared gas analyser (Geotechnical supplies Ltd.). The leachate was analysed monthly for depth in the central well, pH, conductivity, BOD, COD, sulphate, alkalinity, ammonia, nitrate, nitrite, chloride, sodium, calcium, potassium and magnesium. TVA, TON, and phosphate.

Iron and manganese were monitored quarterly and cadmium, zinc, chromium, copper, nickel and lead were analysed annually. At the time of writing the routine analysis of the leachate in the test cells was performed by Bodycote Ltd., Coatbridge, Glasgow (see Section 2). They were employed for the analysis of leachate from October 1998 and latterly throughout the length of the Ammonia Fluxes in Landfills project, contracted by Glasgow Caledonian University.

An automatic weather station provides meteorological data for the site. A further two test cells contain either a mixture of pulverised refuse and sand (27% by volume) (Cell 1), or unpulverised refuse (Cell 2). A further cell containing unpulverised refuse which is not recirculated acted as a control (Cell 4). A report on phase two of the Environment Agency supported Field Trials of Waste Manipulation Techniques project will be submitted for publication in 2002 by the Caledonian Shanks Centre for Waste Management, Glasgow Caledonian University.

Figure 4.1 Scale drawing of the aeration tank.



4.2 Materials and methods

4.2.1 Construction of a batch fed leachate reactor

To provide leachate treatment, an insulated 10m^3 leachate aeration tank (total volume 12.6m^3) was constructed on the surface of the test cell. The glass fibre resin cylindrical tank (Cambrian Plastics Ltd.) was supplied by four 63mm (2") PVC inlet/outlet ports bonded at upper and lower positions at either ends of the tank (Figure 4.1, 4.2). The lid (Camplass, Mid-Glamorgan, Wales) allowed the escape of air through the sides. An end plate allowed the connection of a pneumatic actuated valve (Dryden Aqua Ltd, Bonnyrigg, Scotland). The inlet and exit port were each fitted with an actuated valve. A 'Y' piece was fitted before the inlet valve, which led to another valve and a bypass pipeline, set at an incline, which led directly to the recirculation inlet port. Since both the exit valve and the bypass valve was set on the same operating channel a manual valve (Durapipe 63mm PVC, Regulare) was also fitted to the bypass line to avoid the possible escape of gas or leachate during emptying.

During 2000 an additional manual valve was fitted to enable the tank to remain full while the bypass line was operating. The actuated valves were supplied by an Airmaster Tiger 4/6 compressor. The supply of air to the actuated valves was regulated by a 12V control and timer (Dryden Aqua Ltd., Bonnyrigg, Scotland). This four-channel timer controlled the inlet valve, the exit and bypass valve, the aeration compressor (240V) and leachate pump (240V) (Figure 4.2).

Leachate was pumped into the tank to a total volume of 10m^3, and aerated and nitrified leachate was returned to the landfill leaving three cubic metres residual volume to seed the next treatment. To provide additional surface area for bacterial growth and a greater resilience to fluctuations in temperature and operating performance 90x90x30mm polypropylene bio-medium (Dryden Aqua Ltd., Bonnyrigg, Scotland) was suspended in 8 polypropylene mesh bales (402x402x1004mm) within the tank. The polypropylene mesh (internal diameter 48x48mm) were formed into bales using polypropylene cable ties. Each sack contained 519 bio-media elements.

Pumping was controlled by a timer which opened the pneumatic inlet and outlet actuated valve system. A Rietschie DLT-40 rotary vane air compressor (total capacity 40m^3 h^{-1}) aerated the tank from a submerged ¾" internal diameter perforated PVC pipe. Two brass valves (Europa ¾" pipe valve, Italy) at the compressor and at the top of the tank prevented suck back of leachate through the ¾" braided polyethylene piping. The submerged pipe was held 210mm above the bottom of the tank by four concrete blocks (140x210x440mm) and 10mm galvanised braiding. During the first operational month, two of the concrete blocks were replaced by 5 clay house bricks and polyethylene twine to reduce possible buffering from the concrete. Aeration was adjusted by a 22mm copper 'T-piece', tap and 22mm copper piping held vertically by a post to the same level as the leachate in the tank. The aeration was held between 5-10m^3 h^{-1}.

At the start of the operation of the tank during 2000 a galvanised mesh guard (13mm x 13mm) (Guardman Ltd, Lincs.) was fitted over the exit port within the tank and held firm by a steel jubilee clip. This guard avoided possible blockages to a new 63mm PVC, Regulator valve which was fitted to the outlet port of the tank.

Gas flow and composition were monitored and the leachate sampled regularly. Treatment started when the tank was emptied on 23rd July 1999 and replaced with 2.33 m^3 of leachate from the test cell. To avoid having to remove further leachate in response to the build up of leachate within the test cell the 7.5 m^3 leachate in the tank was removed from site rather than recirculated. The tank was also seeded with 200 l of treated leachate from a treatment lagoon at Glasgow District Council's Summerston landfill site. The leachate treatment tank operated for a total of 5 months during the summer of 1999 over which time four treatments of leachate were made. Following this the test cell was returned to normal recirculation without treatment. Treatment continued in the spring of 2000 but normal recirculation of leachate without treatment was carried out every fortnight when possible, to comply with the other test cells.

4.2.2 Design considerations

Auchencarroch Cell 3 was chosen for this study because it contained the same refuse as was used in previous laboratory experiments performed during the Nitrogen Balances in Landfills project (Burton and Watson-Craik, 1997, 2000). The total volume of the treatment tank was constrained by the wish to maintain the treatment regime firstly, within the normal recirculation of the test cell used, and secondly, with the other test cells at Auchencarroch. Since the constructed system could not be duplicated by another test cell, the experiment was dependent on assessing the previous gas production of the cell prior to leachate treatment and recirculation.

Delays in construction meant that the whole site was recirculated, including Cells 1 and 2, for over a year prior to the operation of the treatment tank on Cell 3. The recirculation of leachate at Auchencarroch ensured the Field Trials of Waste Manipulation Techniques project was maintained before the hand over to Glasgow Caledonian University in 1999. It was initially felt that the sump could yield a limited amount of leachate without running dry and this would limit the recirculation to one hour. This was the case for Cells 1 and 2, but Cell 3 contained high levels of leachate in comparison and so it was possible that recirculation could have been carried out for longer.

The main constraint to recirculating greater amounts lay with the limited void space of the recirculation blanket trench and the design of the central well. The central well was constructed with a perforated pipe which enabled short-circuiting to occur. During recirculation, leachate could travel along the surface or upper layers of the refuse and back into the central well, bypassing the main body of the refuse. This was not apparent to the authors at the instigation of the project but the problem was rectified before the start of the experimental study by digging around the central well of Cell 3 to below 3m, replacing pipe and plugging with a clay surround.

Evidence of leachate short-circuiting was apparent on a couple of occasions prior to remediation but the quantities of leachate entering the central well were limited to a trickle. Attempts to rectify this flaw were made during the original contract of the test cells at Auchencarroch (Envirocentre Ltd., 1998). An extra smaller pipe was placed within the central well. It is possible that it may not have been necessary to remediate Cell 3 since the short-circuiting appeared to be minor, but this may have become more exaggerated as time went on. Cell 1 in the autumn of 2000, during normal recirculation by Glasgow Caledonian University, exhibited a complete failure in the central well and leachate bypassed the refuse completely.

The high leachate levels in Cell 3 at the start of the experimental study, and throughout its operational life, necessitated that some leachate would have to be removed during the project to avoid damage to the cell integrity and possible flooding. Leachate was first partially treated and then removed from the test cell site to the main site following agreement with the Scottish Environmental Protection Agency (SEPA) and the relevant authorities. During the original Waste Manipulation Techniques Project (EnviroCentre Ltd., 1998) approaches were made by the authors to the project managers about placing access ports to allow the refuse to be sampled which would allow the state of decomposition of the waste to be followed. The access ports designed, and constructed by the project's managers were unfortunately often flooded during the operation of the aeration tank and were not available for use. Solid samples were removed prior to the construction of the tank using the project's steel borer/lance but these were sodden, small and mostly of plastic sacks.

As with other field studies, the leachate levels, and therefore the moisture contents between the different test cells at Auchencarroch represent a significant difference, which must be accounted for when the data is interpreted. The difference is more apparent when one considers the depth and volume of the saturated zone since the depth of the fill in these test cells was 5 m following construction.

Figure 4.2 The aeration tank.

4.3 Results

4.3.1 Operation of the batch fed leachate reactor

The first month's operation of the aeration tank is shown in Table 4.1. Recirculation of treated leachate was carried out during the summer of 1999 and 2000.

Table 4.1 Operation of the leachate aeration tank from 27.7.99 to 24.8.99. The tank was filled with 2.33m^3 leachate after 0, 3 and 14 days.

Operation	Day	pH	Nitrate mg NO_3^--N l^{-1}	SD	Ammonia mg $NH_{3/4+}$-N l^{-1}	SD
Start	**0**	**9.12**	**190**	**2.57**	**119**	**3.71**
Fill 1	0	7.27	72.3	0.0	255	6.63
	3	8.32	281	10.1	174	1.94
Fill 2	3	7.32	167	7.37	226	1.94
	14	8.50	302	9.10	172	12.8
Fill 3	14	7.48	220	19.1	229	3.17
Recirculated	**28**	**8.09**	**460**	**14.7**	**0**	
Fill 1	28	7.24	124	4.94	332	13.2

It was possible to alter the time of the second fill from 3 days to 7 days and still maintain complete oxidation of ammonia within one month. The pH of the returning leachate was normally within one pH unit of the leachate in the test cell.

The second years operation was more successful than the first owing the accumulation of bacterial activity and biomass from the first year of operation. Although treatment was achieved during the first year and the recirculation was continued in a monthly cycle, some ammonia was left within the tank on the second recirculation (Table 4.2). Presumably this was because activity was lost after the first cycle as the tank emptied.

Once the transition in activity from suspended bacterial biomass from the seed to an actively growing biofilm on the biomedia elements was made the treatment was more successful and during the third cycle all the ammonia was oxidised within the prescribed period. However, some nitrite persisted, indicating incomplete oxidation by the nitrite oxidising bacteria to nitrate. This contaminated the analysis of nitrate on 19.10.99 but was completed oxidised following emptying in the residual volume to nitrate over 14 days. Nitrite was not found during 2000 and complete oxidation to nitrate was observed (Table 4.3).

Sampling of the aeration tank following filling often proved inaccurate during 2000 as the cylindrical nature of the tank meant that it took a number of hours for the leachate to mix properly throughout the tank. Thus samples taken following filling and aeration were not usually as good an indication of the total treatment as would be achieved when samples were taken a few days before filling. In the later samples, it was usually possible to calculate the expected final nitrate concentration by totalling the ammonia and nitrate concentrations. In laboratory studies

where the same analytical methods were used most ammonia was indeed oxidised to nitrate. Nitrite contamination interfered with some samples towards the end of 1999 (Table 4.2). Variations in the concentrations of ammonia entering the aeration tank during filling also complicated the treatment so final nitrate concentration varied between batches. Based on the laboratory studies, it was not expected that aeration would remove significant concentrations of ammonia by volatilisation. Denitrification was still possible within submerged sediments or biofilms in the aeration tank despite the fact that the tank was nearly saturated with oxygen during aeration.

Phosphate concentration decreased during treatment as it was assimilated and adsorbed by the biomass. The chemical oxygen demand also decreased as the soluble organic compounds were respired and assimilated by the micro-organisms within the aeration tank (Figure 4.3).

Table 4.2 Summary of the treatment of the leachate within the aeration tank after each fill during 1999.

Date	Operation	pH	Nitrate mg NO_3^--N l^{-1}	SD	Ammonia mg $NH_{3/4+}$-N l^{-1}	SD
27.7.99	**[a]Empty**	**9.12**	**190**	**2.57**	**119**	**3.71**
	Fill 1	7.27	72.3	0.0	255	6.63
3.8.99	Fill 2	7.32	167	7.37	226	1.94
10.8.99	Fill 3	7.48	220	19.1	229	3.17
24.8.99	**Empty**	**8.09**	**460**	**14.7**	**0**	
	Fill 1	7.24	124	4.94	332	13.3
31.8.99	Fill 2	7.49	149	3.64	289	22.3
3.9.99	Fill 3	7.69	128	4.27	299	31.4
14.9.99	**Empty**	**8.54**	**173**	**5.61**	**247**	**7.43**
	Fill 1	7.29	61.0	2.57	318	6.34
21.9.99	Fill 2	7.4	183	5.75	264	8.97
28.9.99	Fill 3	7.42	178	5.75	141	3.81
19.10.99	**Empty**	**8.6**	**[b]758**	**12.3**	**0**	

[a]The first treatment was removed from the site. [b]contaminated with nitrite. Total oxidised nitrogen concentration was 350- 380 mg N l^{-1} of which 60 mg N l^{-1} was nitrite.

Table 4.3 Summary of the treatment of the leachate within the aeration tank after each fill during 2000.

Date	Operation	pH	Nitrate mg NO_3^--N l^{-1}	SD	Ammonia mg $NH_{3/4+}$-N l^{-1}	SD
9.5.00	Fill 1	7.33	117	4.60	210	17.9
30.5.00	Fill 2	7.48	211	8.61	143	1.49
6.6.00	Fill 3	7.48	252	1.29	94.4	1.34
20.6.00	**Empty**	**8.5**	**311**	**8.61**	**0**	
	Fill 1	7.23	156	2.13	179	6.34
28.6.00	Fill 2	7.8	158	4.07	151	10.3
4.7.00	Fill 3	7.03	191	7.83	177	3.88
19.7.00	**Empty**	**7.9**	**463**	**7.51**	**0**	
	Fill 1	7.09	187	0.0	268	4.49
25.7.00	Fill 2	7.33	221	17.8	221	4.49
1.8.00	Fill 3	7.46	208	3.34	244	4.49
9.8.00	**Empty**	**7.74**	**373**	**3.34**	**0**	
	Fill 1	6.88	191	3.34	369	13.4
15.8.00	Fill 2	7.97	162	1.29	63.3	1.82
22.8.00	Fill 3	7.88	164	1.29	202	8.97
	***Empty**					

*The last treatment was removed from the site.

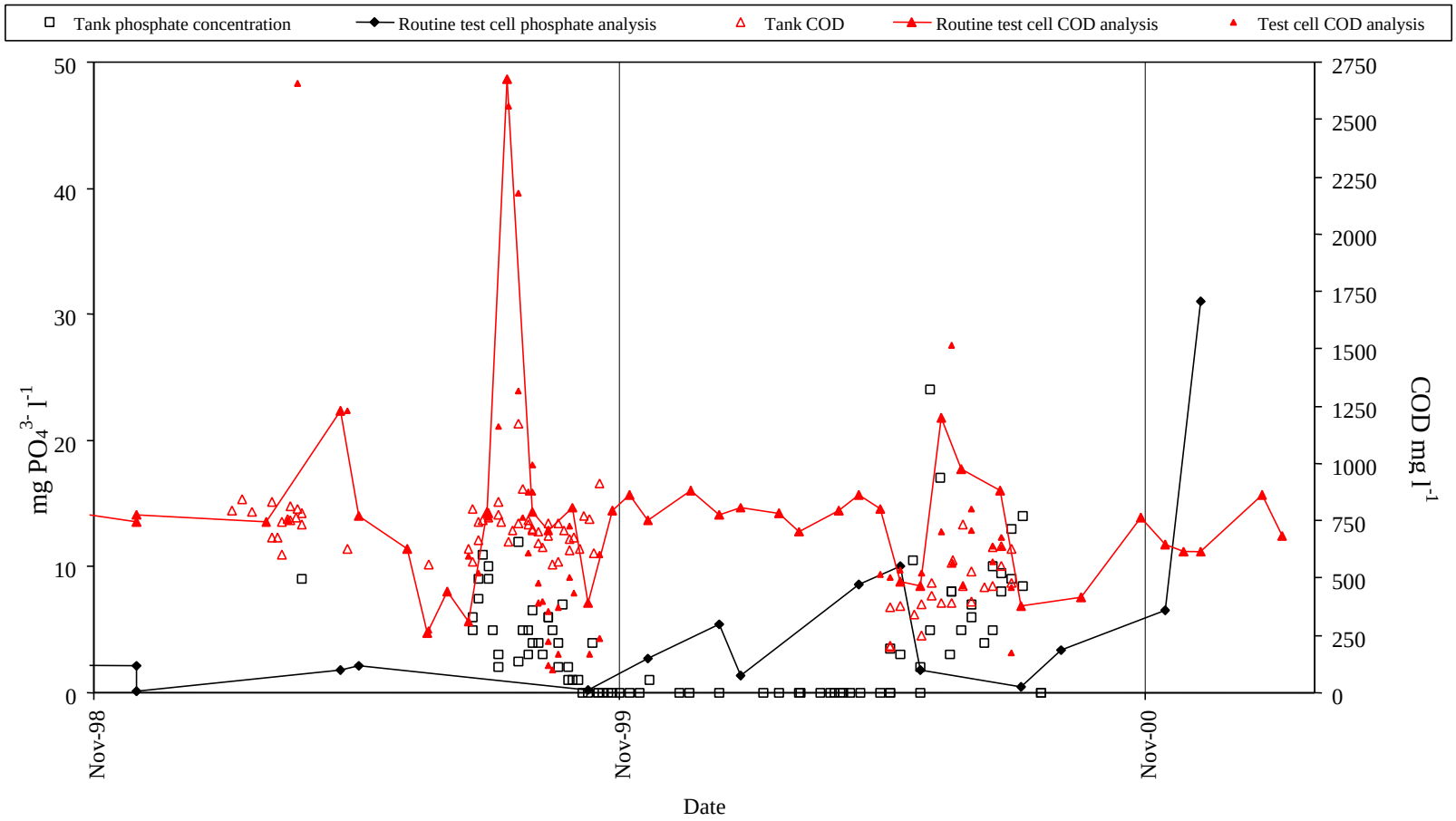


Figure 4.3 COD and soluble phosphate concentrations in the aeration tank and the test cell.

The operation of the leachate aeration tank is shown on Figures 4.3, 4.4, 4.5 and 4.6. Temperature and pH were affected during filling (Figure 4.4, 4.5). Aeration increased the pH of the leachate as the carbon dioxide was driven from the leachate. The increase in pH following filling buffered the aerobic treatment processes, such as the bacterial aerobic respiration of organic compounds and nitrification, both of which lead to acid production and a decrease in pH. Bacterial oxidation of ammonia to nitrite and nitrate proceeded over a one month period. Optimal conditions for nitrification were attained with slightly alkaline leachate pH values. No chemical buffering of the leachate was required, and complete treatment of ammonia was achieved.

When first filled, the pH of the leachate in the aeration tank increased in a manner similar to the treatment of leachate in the laboratory (Figure 3.1). Once the treatment of ammonia to nitrate by nitrification was initiated by inoculation with treated leachate, the increase in the pH value induced by aeration buffered the nitrification process (Table 4.1). Thus the pH following the first addition of leachate was lower by approximately half a pH unit during operation than the pH of the leachate when the tank was first filled and aerated. The pH was further buffered by the 3 m^3 residual volume present in the aeration tank when the system first fills. Further stepwise increases in tank volume during filling help the system maintain the appropriate pH value for rapid nitrification.

Aeration introduced oxygen into the leachate. Using a portable Jenway oxygen electrode it was found that within the bubble stream the leachate was completely saturated with oxygen (approximately 10 mg O_2), based on comparison with saturated distilled water at the same temperature. Moving 300mm from the bubble stream immediately above the hatch at the sampling point the leachate dropped to 95% saturated with oxygen at approximately 150mm into the liquid. Further away the leachate was approximately 90% saturated, dropping to 66-80% after aeration had been stopped for an hour during emptying. The oxygen could fall to between approximately 20 to 69% saturated after filling though this depended where the electrode was placed and how far the stream of pumped leachate entering the tank had to fall from the inlet pipe. When the tank was first aerated following filling traces of carbon dioxide 0.09-1.8% could be analysed in the headspace.

4.3.2 Gas production from the test cell.

Methane concentration in the gas vent decreased during the middle of 1998, recovered slightly to fall in the spring of 1999 and then increased during the later half of 1999 into 2000 (Figure 4.8, 4.9, 4.10). There was no evidence of a decrease in methane concentration owing to recirculation of nitrified leachate.

Methane and carbon dioxide concentrations between 22.4.98 and 15.12.98 were 65.47% (SD 2.56) methane and 34.59 (SD 1.31)% carbon dioxide. Concentrations in 1999 were 61.84% (SD 6.28) methane and 33.41% (SD 4.27) carbon dioxide between 26.1.99 and 14.12.99. The concentrations during 2000 between 11.1.00 and 15.8.00 increased slightly to 66.31% (SD 1.68) methane and 31.86% (SD 1.09) carbon dioxide. Concentrations of methane present in the landfill gas were found to be significantly greater during 2000 than 1999 when compared by Student's t-test ($P<0.001$) with correspondingly less carbon dioxide measured during 2000 than 1999 ($P<0.01$). Whilst there were significant differences between methane concentration during 1998,

1999 and 2000, differences in carbon dioxide concentrations were not very significant between 1998 and the remaining two years (P<0.077) (Table 4.4, 4.5).

Table 4.4 Test cell landfill gas methane concentration during 1998, 1999 and 2000.

Period	% Methane concentration				
	Mean	SD	Maximum	Minimum	Median
22.4.98 - 15.12.98	65.47	2.56	68.8	60.7	65.4
26.1.99 - 14.12.99	61.84	6.28	68.9	39.2	63.6
11.1.00 - 15.8.00	66.31	1.68	70.0	63.8	66.5

Table 4.5 Test cell landfill gas carbon dioxide concentration during 1998, 1999 and 2000.

Period	% Carbon dioxide concentration				
	Mean	SD	Maximum	Minimum	Median
22.4.98 - 15.12.98	34.59	1.31	36.5	32.9	34.1
26.1.99-14.12.99	33.41	4.27	41.5	13.5	34.8
11.1.00-15.8.00	31.86	1.09	34.8	30	31.8

During the period of study the sampled gas pressure was found to be identical to atmospheric pressure. Gas temperatures in the gas vent were also identical to atmospheric temperatures owing to the relative similarity in landfill temperature and the outside environment. Gas temperature measurements performed during gas analysis just prior to the start of the project were believed to be more representative of sample line temperatures rather than gas temperatures and were not continued. The landfill test cell decreased in temperature considerably after construction. There was no accurate method for monitoring gas temperature as it passed through the gas flow meter at or near ambient temperatures. Since this pipe was not insulated the gas was heated or chilled as it passed from the gas flow meter to the vent. The Inner I temperature probe may have given the best assessment of gas temperatures since it was physically very close to the gas flow meter. Propane gas ejected from the gas flow meter would also have affected the gas temperature. Against a background of a low flow rate and air in the gas vent it was possible to analyse propane in the sampled gas using the infrared gas analyser after propane was released into the gas pipe during flow measurement.

During 1998 the gas flow meter operated only intermittently despite servicing, and little gas flow data was made available to the authors during 1998. In 1999 and 2000 the gas flow meter operated more regularly and reliably. There was no immediate discernible decrease in landfill gas flow during the recirculation of nitrified leachate during 1999 and 2000. When recorded, gas flow increased during 1999 and 2000. This was observed despite a fall in landfill temperature. There was no evidence of a change in the pattern in the gas concentrations in the days following the emptying of the aeration tank. Methane and carbon dioxide concentrations and the total gas analysed were similar throughout this period. Hydrogen gas was present only in trace amounts.

Gas concentrations were measured and recorded prior to recirculation. The gas concentrations immediately following recirculation were tested during operation of the tank in 1999 and the winter of 1999 during normal recirculation. Methane and carbon dioxide concentrations fell immediately after recirculation but this was due to air being drawn back down the vent. Methane concentrations recovered after several hours in a similar manner to routine recirculation without leachate treatment. Since the design of the test cell places the recirculation blanket and the gas collection system close to one another, recirculation will affect the production of gas.

The remoteness of the landfill site and cost considerations during the original field trials experiment meant that it was necessary to move a single pump powered by a generator from test cell to test cell, and the central well of each test cell was left open during recirculation. Thus some landfill gas by-passed the gas flow meter during recirculation venting through the central well. Thus a certain amount of gas was not accounted for during the recirculation of the test cells which may or may not be proportional to the amount of leachate recirculated. The gas lost, if proportional to the amount recirculated, will not be significant compared to the overall gas volumes released by the gas collection system. The time during which the central well was open was recorded. The gas flow was reduced to zero during the later half of the recirculation period, presumably when the blanket was at or near full capacity. Air was also sucked back into the vent so that the concentrations of oxygen present resembled that of the ambient air surrounding the test cells.

4.3.3 Leachate analysis from the test cell

The authors, Envirocentre Ltd. and Glasgow Caledonian University performed leachate analyses from Auchencarroch Test Cell 3 (see Appendix 2). Assembling these data from the various contributors and their corresponding DoE and EA funded contracts produced a complete historical record of leachate quality from Cell 3. These were referred to as 'routine analyses' and relate to the regular and routine analysis of the leachate (Appendix 2). Some of these data may have been reported previously. The routine analysis data is displayed in Appendix 2. Other analyses were carried out by the authors specifically for the Ammonia Fluxes in Landfills project. These, when possible, were displayed separately in the figures as they used a consistent method of analysis. They can, as in the case of Figure 4.3 and 4.11, be interchangeable, since they often measure the same parameter.

The removal of ammonia from landfill leachate by recirculation of treated leachate

Leachate ammonia concentrations within the landfill during 1999 decreased during the months that treated leachate was recirculated (Figure 4.9, 4.10, 4.11). Periodic additions of leachate to the tank meant that some leachate was at the start contaminated by rainwater infiltration. Ammonia concentration decreased but it was sometimes associated with decreases in conductivity and chloride concentration suggesting rainwater infiltration (Figure 4.12, 4.13). This was apparent on the occasions when manual sampling was undertaken by Waterra tubing to see if nitrate was getting into the central when the tank was emptied prior to filling. This effect was reduced during 2000 when untreated leachate was recirculated, thus mixing the leachate within the cell. Concentrations of ammonia returned to levels representative of the start of the study but decreased when the aeration tank was operated again in 2000. The lowest recorded ammonia concentrations occurred in the autumn of 1999 and 2000 at the end of the two summer treatment

and recirculation periods. When the treatment was stopped in the autumn of 2000, further recirculation without treatment during 2001 mixed the leachate but the ammonia concentration did not increase to the levels prior to the start of the experiment. There was a reduction in test cell ammonia concentration by approximately 25% between the start of the experimental period in 1998 and the concentrations analysed in 2001 when normal recirculation without treatment had been resumed. Thus the operation of the treatment system led to a net reduction in the ammonia concentrations observed in the landfill.

Leachate nitrate concentrations

Although the returning leachate that was returned to the landfill following the emptying of the aeration tank contained nitrate, nitrate was rarely identified during the routine analysis of leachate. Nitrate and nitrite were generally absent from the routine analysis data except during the first 6 months and on 2.3.99, 22.6.99 and 3.8.99. The later readings were low and contradicted chemical analysis on site which at this stage showed no forms of oxidised nitrogen present. The early readings were unusually high and possibly a feature of the analysis method chosen. In these early stages, electrodes were used by the contractor for nitrate measurement, and these measurements were unlikely to be very accurate. The authors have doubt in the accuracy of the early nitrite and nitrate data provided by the contractors, by EnviroCentre Ltd. That said, there was a steady decrease in the concentration of nitrate and nitrite as they were consumed by the anaerobic decomposition processes.

Nitrate concentrations following the emptying of the aeration tank

Attempts were made following the emptying of the treatment tank to measure nitrate in the central well. During the operation of the aeration tank between 27.7.99 and 19.10.99, 74.6 (SD 2.57) mg NO_3^--N l^{-1} was identified three days after the emptying and filling of the aeration tank on 24.8.99, though nitrate could not be found in the central well immediately following the emptying of the tank. When the leachate was sampled again on 31.8.99 nitrate was absent from the leachate. Nitrate was not analysed in the leachate again until 22.10.99 when 126.56 (SD 3.64) mg NO_3^--N l^{-1} was analysed following the emptying of the tank on 19.10.99. This decreased to 35.0 (SD 0.257) when the central well was sampled on 26.10.99, and no nitrate could be found on 29.10.99. No nitrate was found to enter the central well immediately following emptying of the tank.

During the operation of the aeration tank from 9.5.00 to 9.8.00, 50.8 (SD 1.29) mg NO_3^--N l^{-1} was analysed in the central well following the emptying and part filling of the tank on 20.6.00. This concentration had increased to 145 (SD 1.11) mg NO_3^--N l^{-1} when the central well was again sampled on 21.6.00. The concentration fell to 5.65 (SD 0.257) mg NO_3^--N l^{-1} on 28.6.00. Nitrate concentrations of 5.87 (SD 0.00) and 3.84 (SD 1.80) mg NO_3^--N l^{-1} were identified on 19.7.00 and 9.8.00 respectively following the emptying and part filling of the aeration tank. No nitrate was present in the central well immediately following the emptying of the tank, and only with further movement of leachate during pumping did nitrate enter the central well.

Leachate phosphate concentrations

Phosphate was present in the landfill leachate throughout the study. Between the start of the Field Trials of Waste Manipulation Techniques project on 22.11.95 and the last analysed sample on 6.2.01 there were between 0.12 and 31.0 mg PO_4^{3-} l^{-1} in the leachate from the test cell. From 23.1.98, when samples were first analysed for the Ammonia Fluxes in Landfills Project concentrations and 6.2.01, concentrations were again found to be between 0.12 and 31.0 mg PO_4^{3-} l^{-1} (see Appendix 2).

Leachate organic content

The chemical oxygen demand (COD) gradually decreased from the time of construction to the final sample taken on 6.2.01 (Appendix 2). Some variation was observed but the overall trend was a decrease in the COD, signalling a reduction in the organic content of the leachate and the reduction in the observed concentration of volatile fatty acids such as acetate (Appendix 2). The reduction in COD is typical of the transition from acetogenic to methanogenic leachates. Volatile fatty acids such as acetate produced by anaerobic bacterial fermentation are reduced further, to methane and carbon dioxide, by methanogenic bacteria with a reduction in the COD of the leachate. The concentrations of COD were in turn related to the biochemical oxygen demand (BOD) which also shared a decrease throughout the sample period. The two methods both measure the oxidisable organic components in leachate. Total organic carbon (TOC) and total volatile acids which are also measurements of the organic load of leachate shared similar decreases reflecting a decrease in organic compound within the leachate (Appendix 2).

The operation of the aeration tank from 27.7.99 when the leachate was treated and oxidised may have led to a further reduction in the COD of the leachate within the test cell. The COD of the leachate within the tank was reduced by bacterial respiration and activity during this aeration period and probably led to a reduction in the COD of the landfill leachate through treatment and dilution. Cells 1 and 2 at the Auchencarroch Test Cell facility did not show a significant reduction in their COD during the comparable period. The COD concentration from Cell 3 on 23.4.99 prior to the recirculation of treated leachate was approximately the same as that observed in Cells 1 and 2 in November 2000 (data not shown) though both these cells had slightly lower moisture contents. There was some evidence that the treatment of leachate led to a reduction in landfill COD from the landfill. However, during 1998 there was a steady trend in the reduction of COD observed prior to commencing the recirculation of treated leachate.

A range of volatile fatty acids (C_2-C_5) such as acetate, proprionate, iso-butyrate, butyrate, iso-valerate and valerate was evident during 1995-1996, a feature of the acetogenic stage of refuse decomposition. These organic acids concentrations were not present in the routine analysis of leachate by the end of 1998 (Appendix 2). The routine analysis data suggested that from 1998 that the intermediates from the bacterial hydrolysis and fermentation of the waste were consumed as soon as they were formed Appendix 1). The rate-limiting step in refuse decomposition therefore changed from organic acid consumption to the hydrolysis of the waste.

Volatile fatty acids concentrations more representative of acetogenic leachates were determined on 10.8.99, 24.8.99, 3.9.99 and 20.6.00 (Table 4.6). These also corresponded with high COD concentrations observed in the leachate (Figure 4.3). Volatile fatty acids were undoubtedly

present on 23.3.99 and 27.4.99 when high COD values were observed, but these samples were taken prior to the operation of the tank in 1999 have not yet been analysed for volatile fatty acids. No significant concentrations of volatile fatty acids were measured when the leachate from the central well was periodically sampled for pH, conductivity, nitrate and ammonia and these concur with routine analysis (Appendix 2). There is evidence that during the project some parts of the test cell were indeed acetogenic and that conditions were not uniform throughout the whole of the test cell. These volatile fatty acid concentrations were not apparent or were missed during the less frequent routine analysis of the landfill.

The recirculation blanket on the test cell distributed the leachate during recirculation and emptying of the aeration tank, and some impermeable or dry zones would presumably then be flushed giving periodic increases in volatile fatty acids. It is possible that the operation of the tank directly contributed to volatile fatty acid production but since the presence of volatile fatty acids was not necessarily related to the emptying of the tank it is unlikely. The presence of low but quantifiable hexanoic acid concentrations in some of the samples was unusual since it has only ever been found in one batch of 2-3 month old pulverised refuse in over half a dozen visits to the Shewalton landfill site. Its presence may indicate the decomposition of fat.

Table 4.6 Test cells leachate samples containing volatile fatty acids during 1998-2000. Concentrations represent the means of duplicate or triplicate analysis.

Date	acetate mg l^{-1}	propionate mg l^{-1}	isobutyrate mg l^{-1}	butyrate mg l^{-1}	isovalerate mg l^{-1}	valerate mg l^{-1}	hexanoate mg l^{-1}
10/08/99	147.5	39.4	20.6	42.3	20.4	23.6	45.5
24/08/99	623.3	108.5	41.6	41.4	35.3	13.4	25.8
24/08/99	290.6	47.2	11.4	12.8	12.8	4.80	22.1
03/09/99	88.9	9.63	6.87	6.48	4.29	2.08	10.9
20/06/00	248.0	40.1	26.2	55.0	21.2	13.0	40.5

General leachate chemistry

Appendix 2 shows the routine analysis of the leachate from the test cell from the start of the Field Trials of Waste Manipulation Techniques project on 22.11.95 to the last analysed sample on 6.2.01. The last recorded entry for the Field Trials of Waste Manipulation Techniques project was 20.8.97 (EnviroCentre Ltd., 1998). The Ammonia Fluxes in Landfills Project continued to the end of 2000. During 1998 the samples were removed from the test cells and analysed by EnviroCentre Ltd. Samples were at the same time removed by the authors and frozen for reference. These were defrosted over two days in the authors' laboratory and then sent by the Caledonian Shanks Centre for Waste Management, Glasgow Caledonian University for analysis by Bodycote Ltd. One sample taken on 2.12.98 was sent directly by the authors for analysis by Bodycote Ltd. and also frozen for analysis with the other frozen samples from 1998.

Chloride concentrations on 2.12.98 were affected by the freezing and thawing process and were significantly lower than the fresh sample taken on 2.12.98. Calcium and iron were similarly affected but ammonia, because of its high solubility, appeared unaffected. This method of sample preparation and storage was used successfully during the previous Nitrogen Balances in Landfills project during studies on ammonia flushing from refuse columns.

Sodium, potassium and calcium concentrations fluctuated from 1995 to 2000. Their concentrations were related to the conductivity. Calcium concentrations were greatly affected by the storage and freezing of samples during 1998, and like chloride concentrations, these analyses were considered inaccurate. Similar concentrations of sodium, potassium and calcium were observed at the beginning and the end of the analysis period respectively (Appendix 2). Their persistence in landfill leachate is a feature of their high solubility and their ubiquitous presence in the environment and municipal solid waste.

Low concentrations of metal cations such as cadmium, chromium, nickel, zinc, copper and lead were measured in the landfill leachate. Such concentrations are as much a feature of the low solubility of the hydroxide and carbonates of these elements as their actual concentrations in municipal solid waste. Manganese was found in higher concentrations in the first sample on 22.11.95 when 26 mg l^{-1} was observed. Concentrations afterwards were significantly lower at <2 mg l^{-1}. Low concentrations of sulphate were observed during the routine analysis of leachate (Appendix 2). Sulphate was observed in higher concentrations in leachate during the operation of the aeration tank. Laboratory analysis during treatment studies in 1998-1999 showed the leachate to be extremely low in sulphate (data not shown).

Th pH value of the landfill remained constant at approximately pH 7 (Figure 4.6). Unusually for a landfill the first recorded sample was at pH 6.8, despite the concentrations of C_2-C_5 volatile fatty acids, so the acetogenic period of refuse decomposition was extremely short following capping. The pH values were probably not sufficiently acidic during the acetogenic phase for methanogenesis to be severely inhibited. Methanogenesis was consequently rapidly initiated which buffered the organic acid producing stage associated with the start of refuse decomposition. This effect was likely to be a feature of the pulverisation of the waste which will encourage the decomposition processes. The refuse was also capped relatively rapidly in comparison to normal landfills. This will have excluded air and reduced the potential for bacterial respiration to carbon dioxide which reduces the pH. Low redox potentials will have been reached quickly, allowing the alkali producing processes such as methanogenesis to proliferate.

4.3.4 Landfill temperature

The temperature of the landfill slowly decreased during the period of study. The shallowest Inner I and Outer I thermocouples were more susceptible to outside temperature variations. Landfill temperature was similar to the temperature of the leachate during treatment in the aeration tank (Figure 4.14, 4.15).

4.3.5 Test cell leachate level

Total depth of the test cell was 8m from the lip of the central well to the bottom of the sump. The cap depth was 2m on construction. Leachate levels in the central well at the start of 1999 were approximately 5m dropping to 4.4m following removal of 7.5m^3 of leachate. Operational depth during treatment was approximately 4m. The refuse depth in the test cell design was 5m, though the depth of the central well exceeded this. The authors were not aware of the high leachate levels within the test cell at the instigation of the research project. In order to maintain the test cell safely with similar leachate levels, leachate was initially removed from the site following

treatment and again at the end of the experiment. The design of the test cell, its flat level cap which allowed the pooling of water following rainfall, and the very high moisture content in the test cell before capping led to unsuitable leachate levels and problems for the future management of the site. The wet Dano drum process will have also contributed to the high leachate levels since there was limited absorptive capacity left in the waste at moisture contents of above 65% when the refuse was emplaced. Gas production will have been reduced owing to the limited void space afforded by the high leachate levels. The decomposition processes will also have led to the production of water. The Auchencarroch test cells were also sited in a region of extremely high rainfall. The other 3 test cells contained much less leachate, and during 1998 their leachate levels were lower.

4.3.6 Removal of leachate from the test cell

Owing to concerns about the high leachate levels discovered in the test cells at the start of the study, which may have affected the project, the first treatment was removed from the landfill site. A total of 7.5 m^3 was removed on 23.7.99 and 10m^3 of treated leachate was left in the aeration tank at the end of the study for disposal off site. The total volume of leachate removed from the landfill test cell was 17.5 m^3.

4.3.7 Comparison of Test Cell 3 to the remaining Auchencarroch test cells

The Auchencarroch Test Cell Facility comprised 4 test cells. Gas flow rates from Cell 1, that contained pulverised refuse, blended with inert sand (27% by volume), were successfully measured with Cell 3 until winter 1999-2000 when recorded flows were extremely high and the gas flow meter was adjusted for 2000. Allowing for a large gap between 1998 and 2000 the gas flow rates achieved were slightly greater than that of Cell 3 possibly due to the greater void space afforded by the lower leachate levels. Ignoring the winter 1999-2000 period, there was a steady increase in gas flow from Cell 1.

A lithium tracer study was carried out during July 2000 which will yield information on the distribution of leachate during recirculation and will be described in detail in the Auchencarroch Test Cells Phase II report prepared by Glasgow Caledonian University. Unfortunately, Cell 1 became flooded so it is not possible to compare ammonia levels in 2001. Cell 1 and Cell 3 had similar initial ammonia concentrations after construction until May 1999 when the operation of the aeration tank and the treatment began to interfere with leachate collection and reduce the ammonia concentration in the central well. Concentrations of ammonia in Cell 1 remained at approximately 500 mg $NH_{3/4}^{+}$-N l^{-1} until July 2000. Thus the treatment of leachate on Cell 3 had the effect of reducing ammonia concentration in comparison to Cell 1. Cell 2, which contained unpulverised refuse, had a similar concentration of ammonia to Cell 1. Final recorded ammonia concentrations for Cell 2 on 31.10.00, 17.11.00, 30.11.00, 21.12.00, 23.1.01 and 6.2.01 were 437, 404, 494, 353, 444 and 394 mg $NH_{3/4}^{+}$-N l^{-1} respectively. These compared to 302, 282, 342, 259, 311 and 290 mg $NH_{3/4}^{+}$-N l^{-1} for Cell 3 (Caledonian Shanks Centre for Waste Management, Glasgow Caledonian University pers. com.). Gas flow from Cell 1 was slightly greater than that achieved by Cell 3.

Cell 4 contained unpulverised refuse and its leachate was not recirculated. It had some of the lowest recorded gas flows and the lowest mean recorded ammonia concentration during 1995-2000 at 187 (SD 53.3) mg $NH_{3/4}^{+}$-N l^{-1} with limited well purging. A full report on the Auchencarroch Test Cell facility will be published by the Caledonian Shanks Centre for Waste Management, Glasgow Caledonian University.

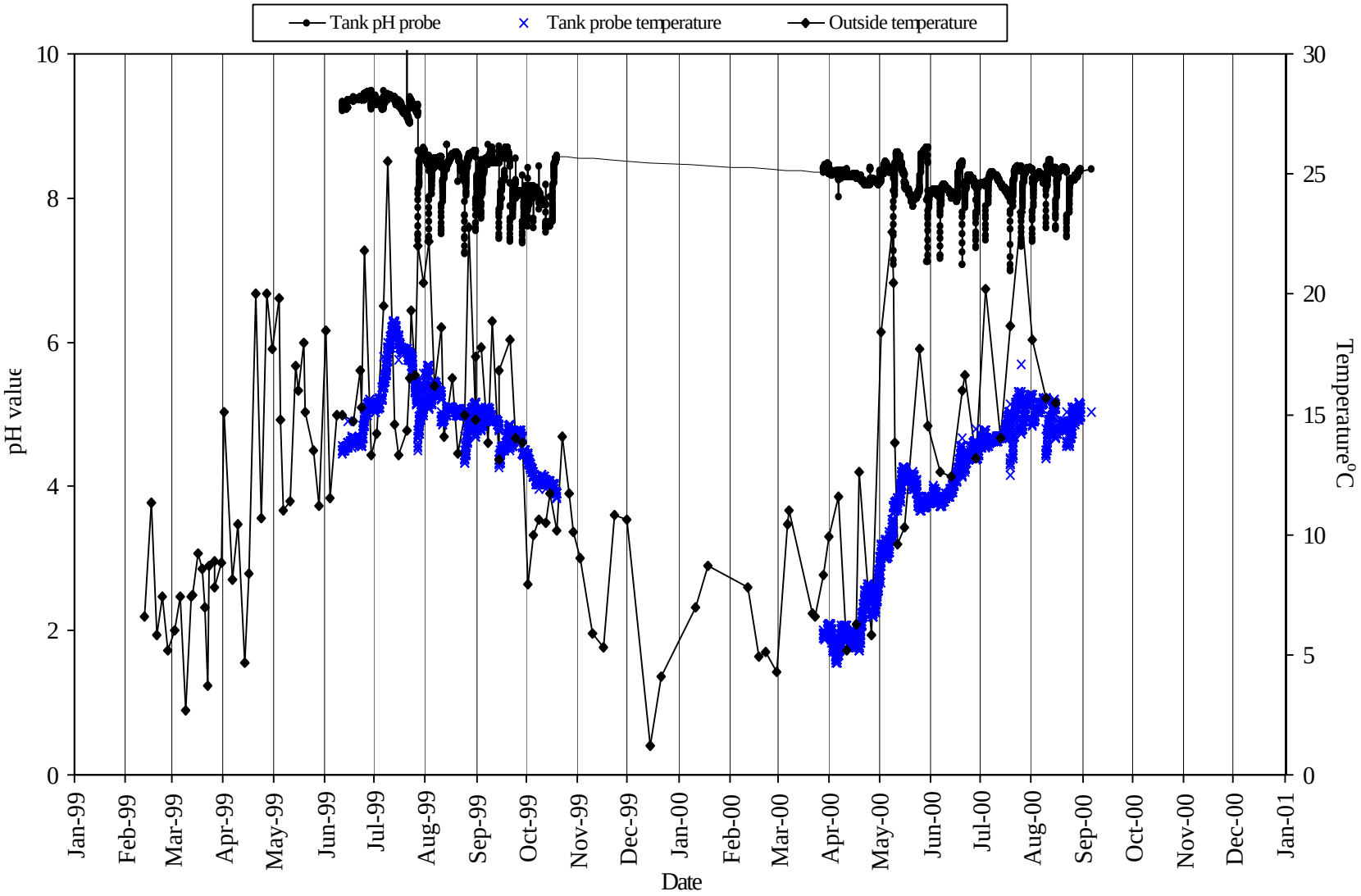


Figure 4.4 Aeration tank probe pH value, temperature and outside temperature.

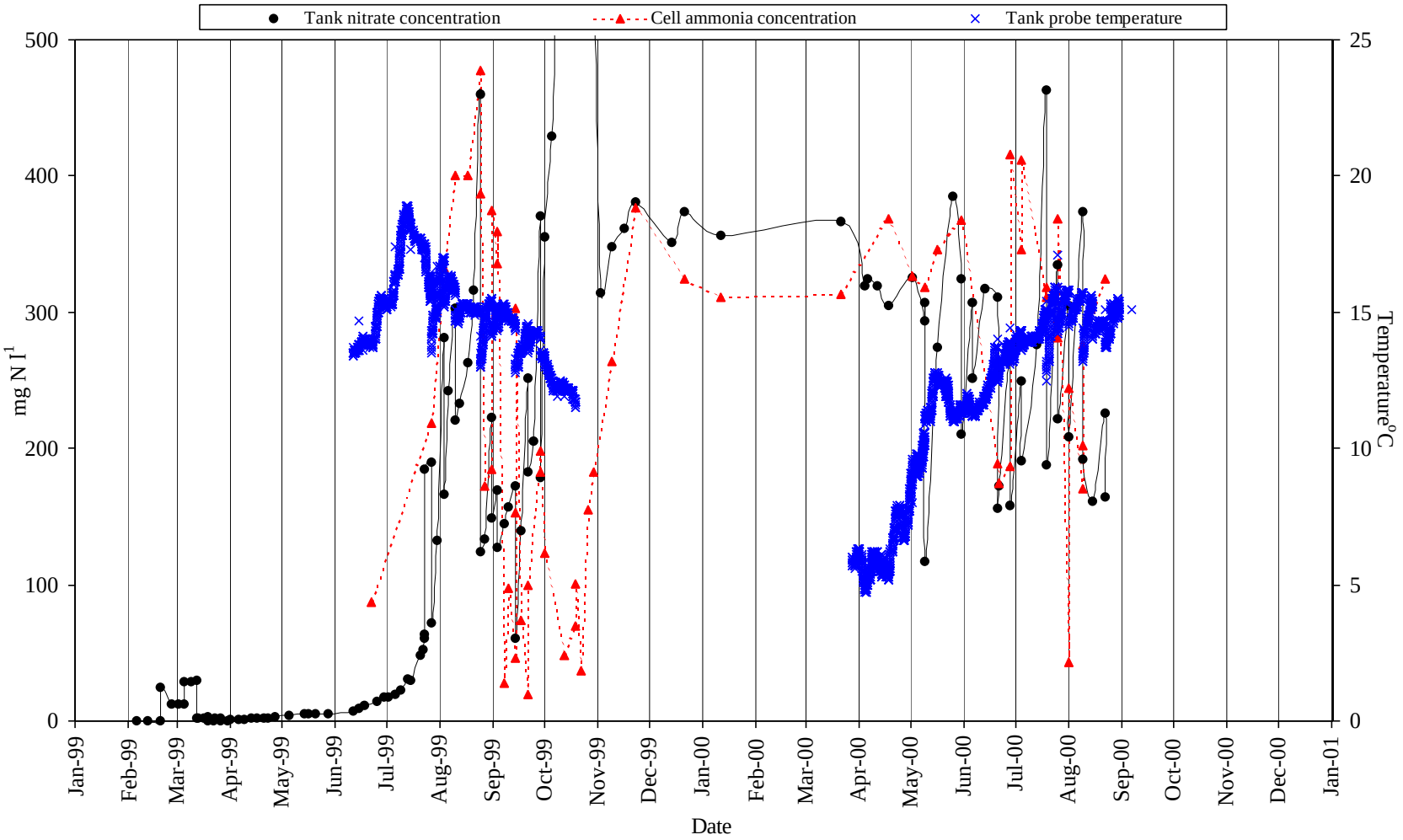


Figure 4.5 Aeration tank temperature, nitrate concentration and test cell ammonia concentration.

Figure 4.6 Aeration tank leachate temperature, pH value, external temperature (12pm-1pm) and routinely analysed test cell pH value.

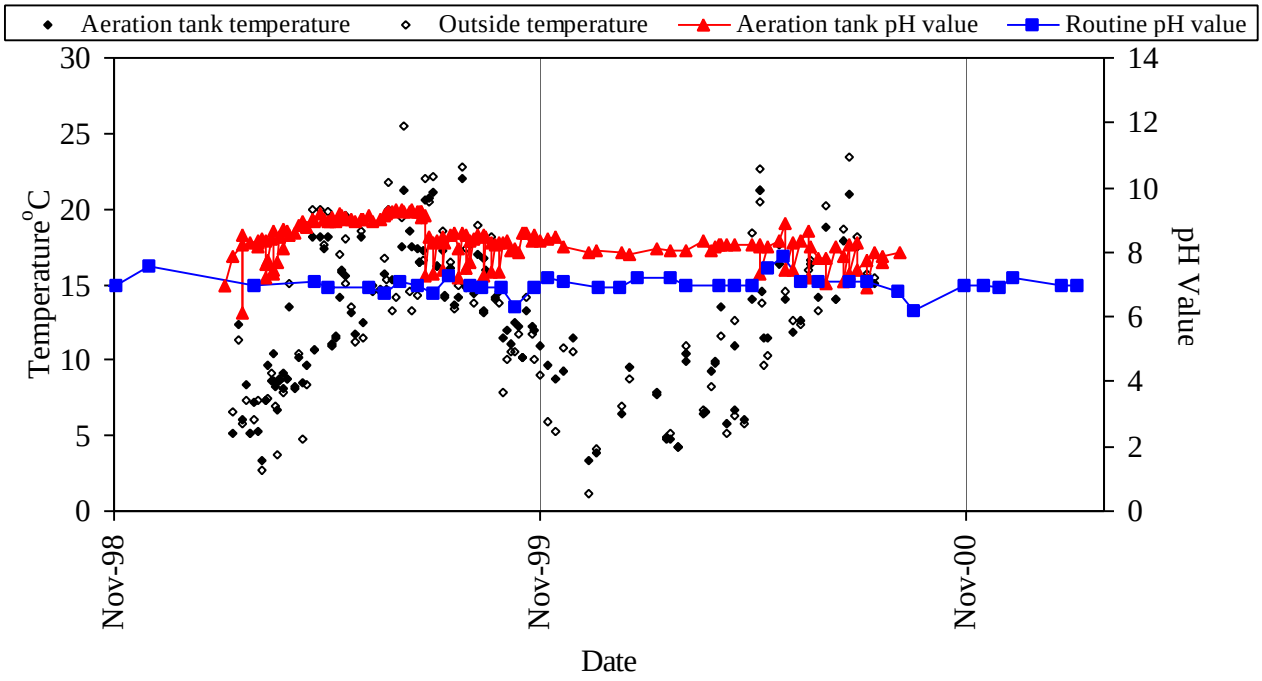


Figure 4.7 Test cell ammonia concentrations taken during the Ammonia Fluxes in Landfills project and routinely analysed chloride concentrations.

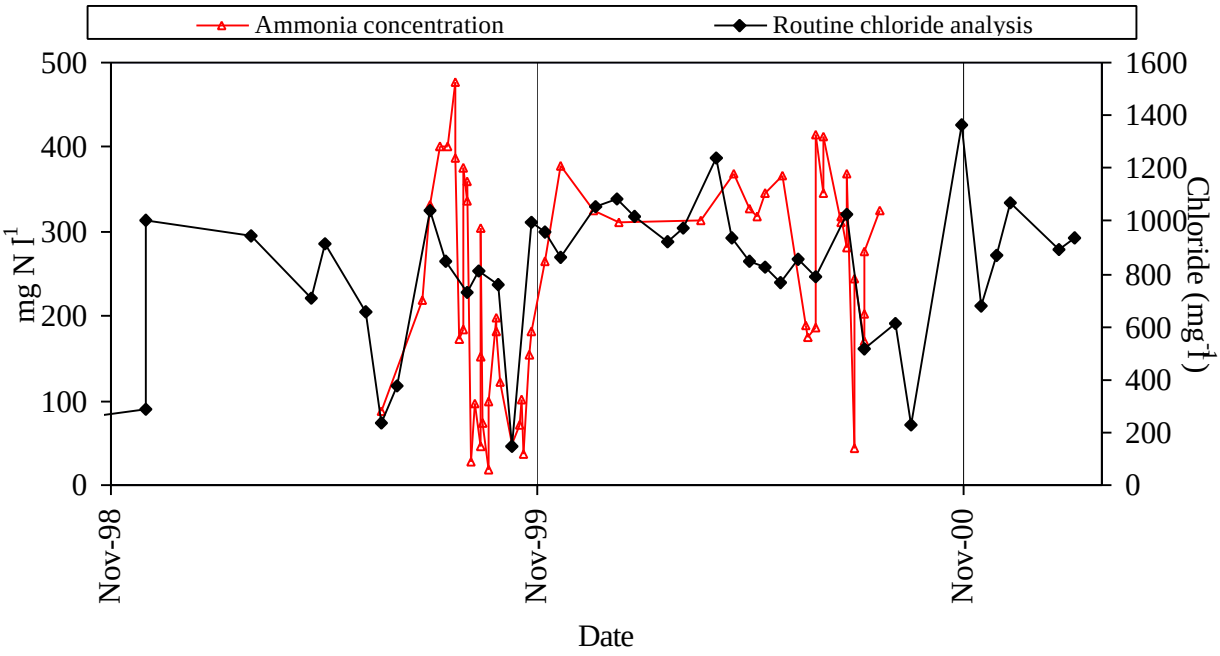


Figure 4.8 Gas flow and gas quality from the test cell. The trendline was calculated for gas flow from 1995 to 2001.

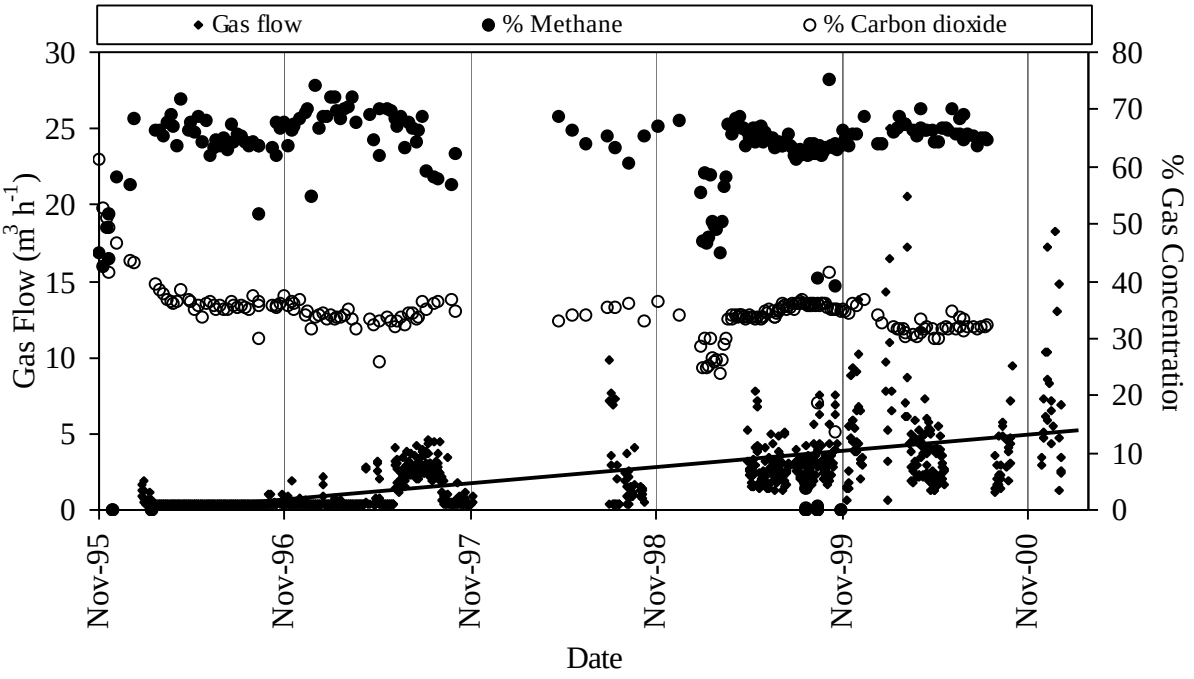


Figure 4.9 Gas quality, routine ammonia concentrations and ammonia analyses from the test cell. The trendline was calculated using the routine analysis of ammonia from 1995 to 2001.

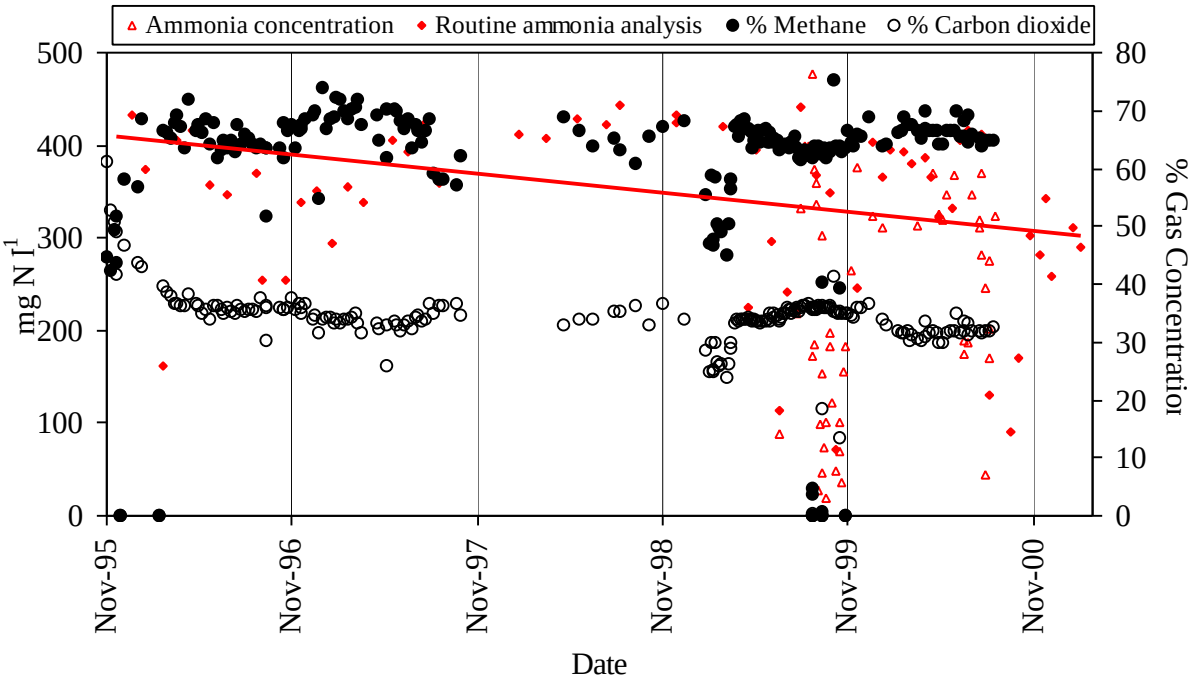


Figure 4.10 Routine test cell ammonia and gas concentrations recorded during the Ammonia Fluxes in Landfills project 1997-2001.

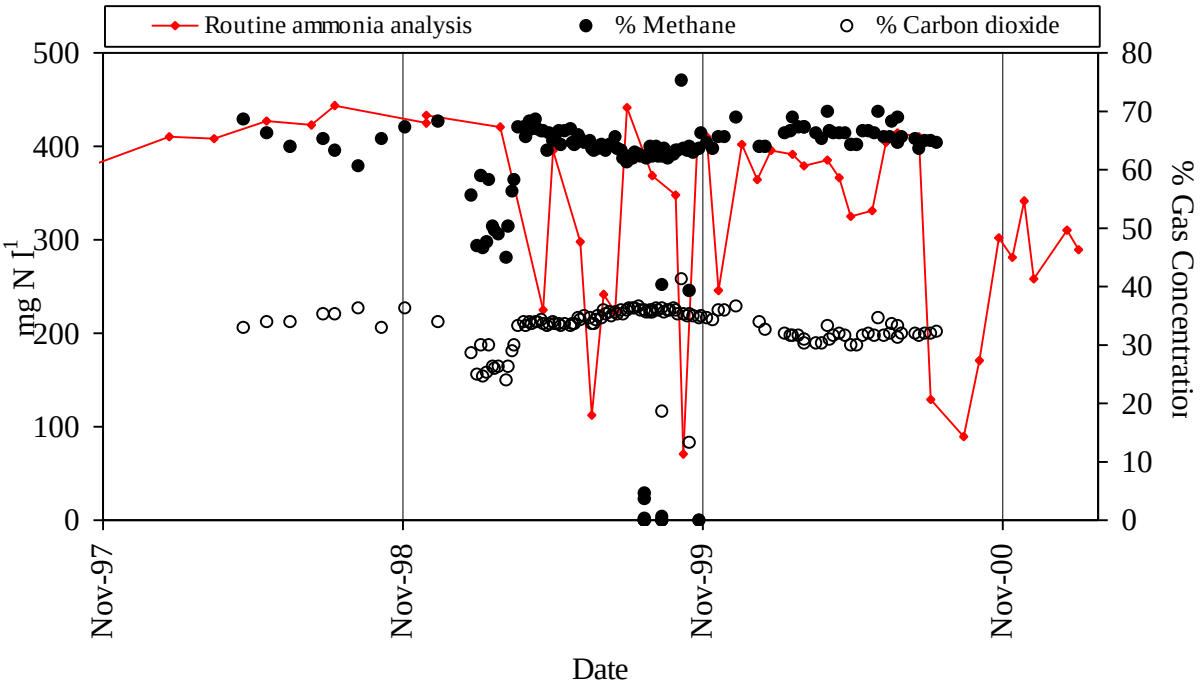


Figure 4.11 Test cell leachate ammonia concentrations and routine ammonia concentrations recorded during the Ammonia Fluxes in Landfills project 1997-2001.

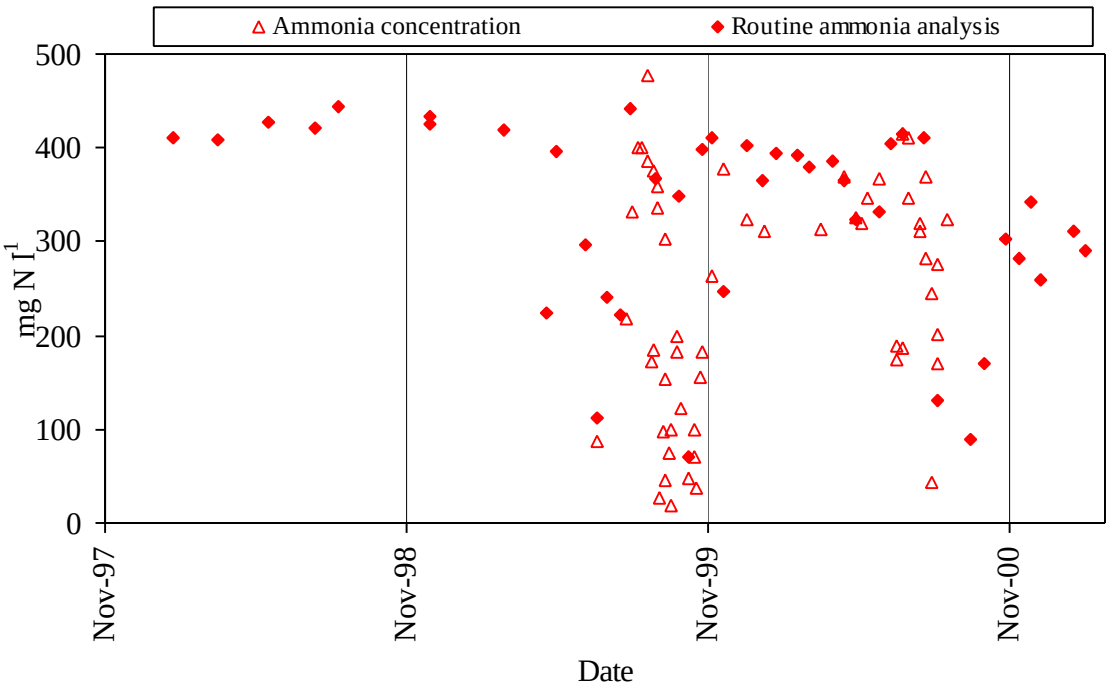


Figure 4.12 Test cell ammonia concentration, routine ammonia analysis and conductivity.

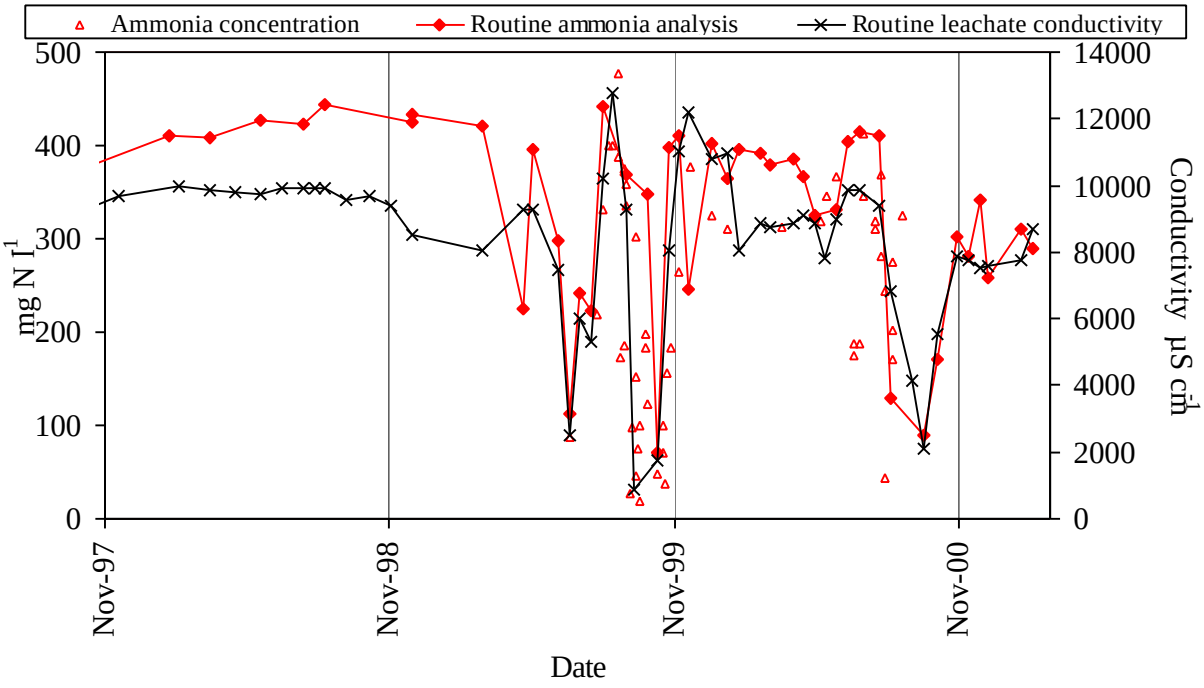


Figure 4.13 Test cell ammonia concentration, routine ammonia analysis and routine chloride analysis (unfrozen).

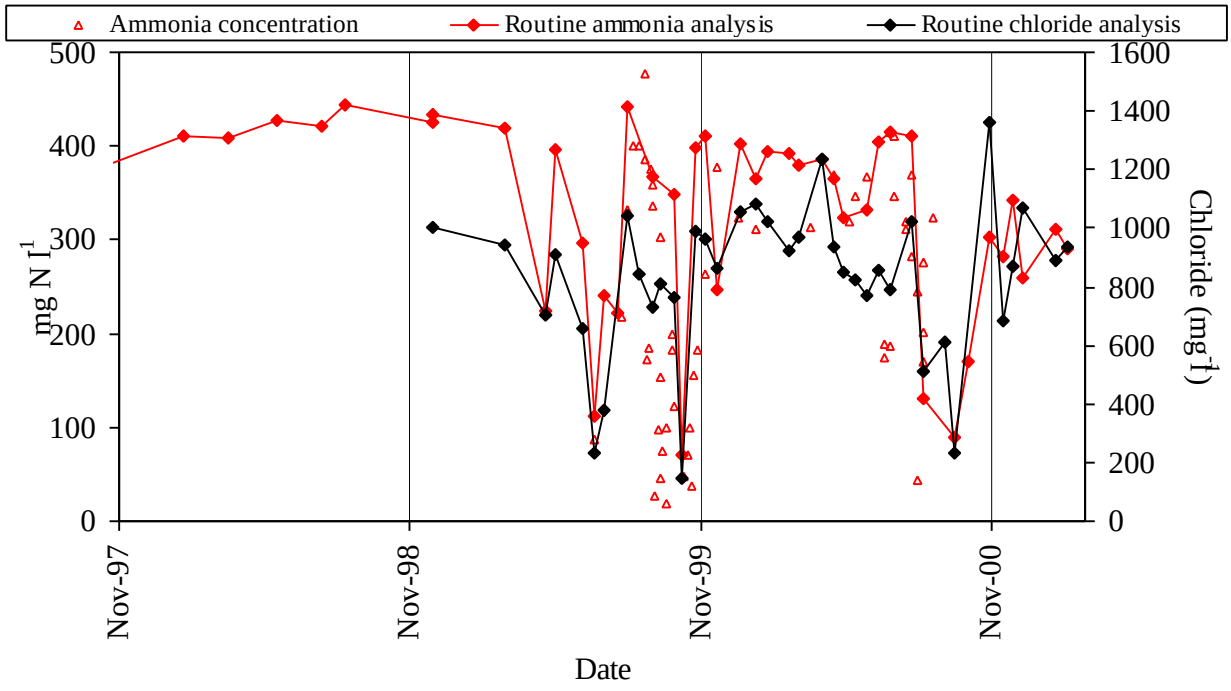


Figure 4.14 Test cell methane concentrations and the 'inner' array of temperature probes. The probes are arranged in order of depth with 'inner I' the shallowest.

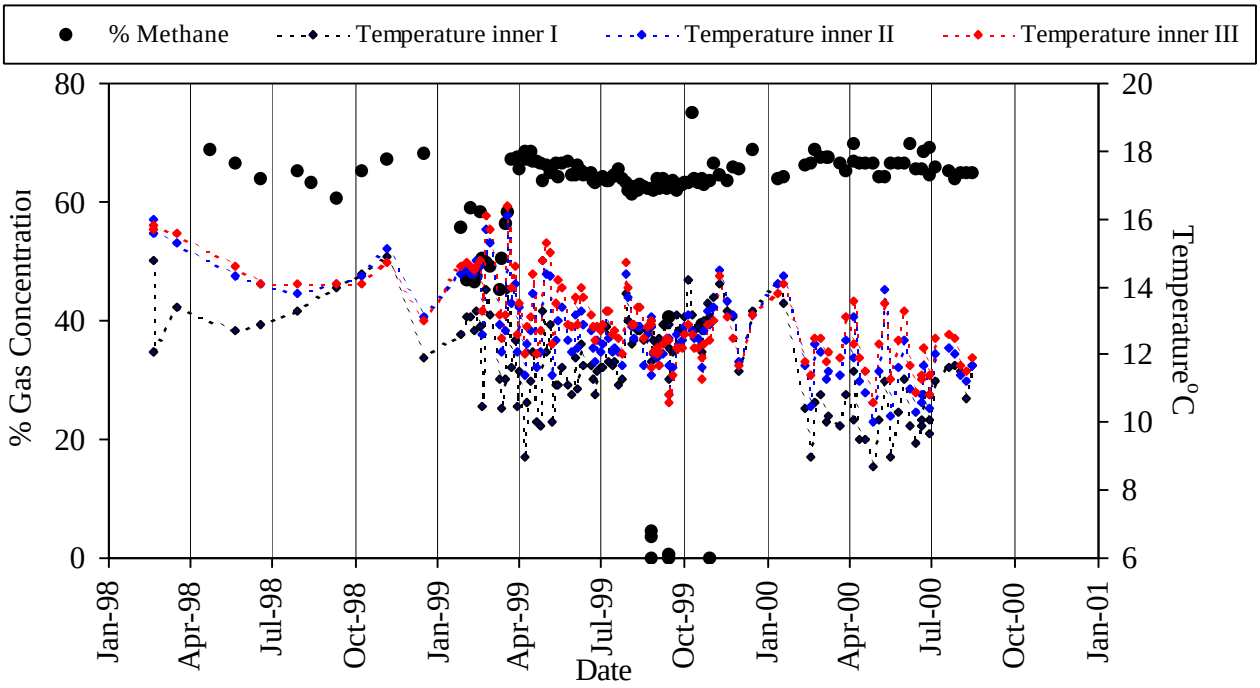
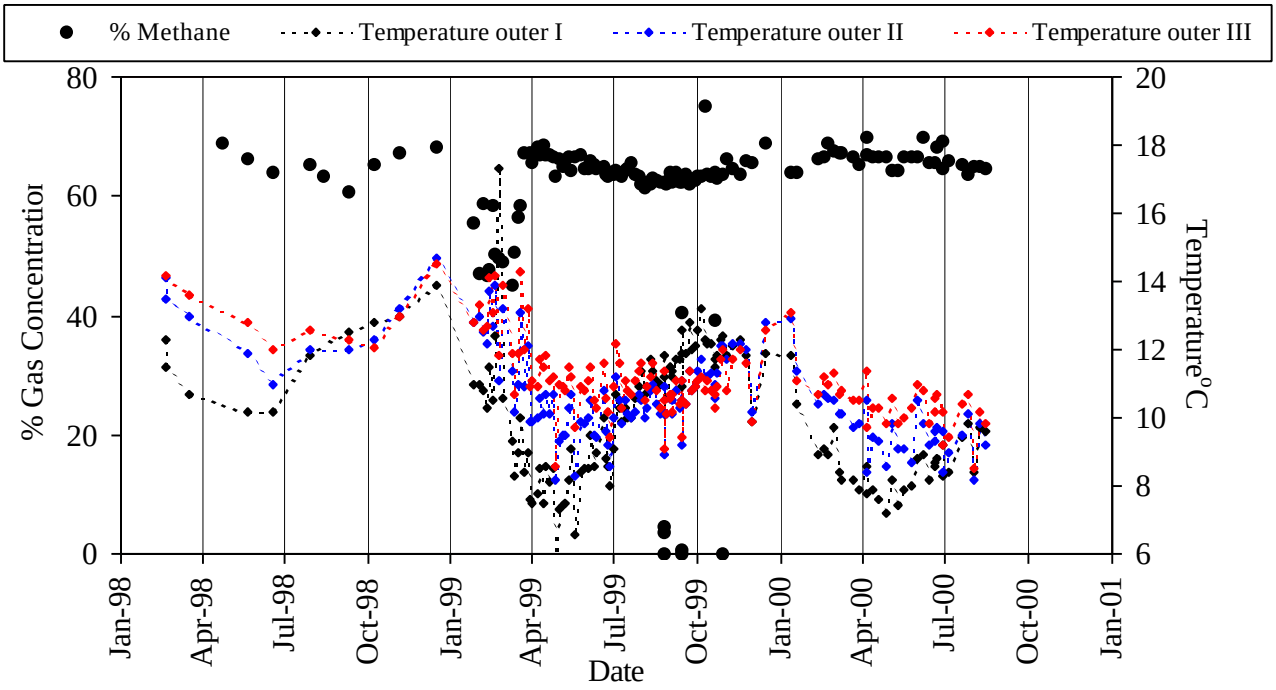


Figure 4.15 Test cell methane concentrations and the 'outer' array of temperature probes. The probes are arranged in order of depth with 'outer I' the shallowest.



5. DISCUSSION

5.1 Aeration tank performance

The aeration tank was operated in a batch fed sequence primarily to allow the recirculation to continue at similar levels prior to the start of the recirculation of treated leachate. Continuous recirculation was not felt to be a suitable option in that it may promote channelling through limited portions of the waste. Recirculation of large volumes allowed the distribution of leachate throughout the gravel recirculation blanket and reproduced the operating conditions of the test cell prior to the start of the experiment.

The ease, at which the leachate from the test cell could be nitrified, both in the laboratory and in field studies, without pH control, is to be noted. The pulverised MSW within the landfill test cell after approximately three years recirculation, did not present any problems for leachate treatment. The leachate was saturated with carbon dioxide, which when aerated, was driven off, altering the equilibrium to moderately alkaline conditions. Following treatment the recirculated leachate had a pH value of between 8.09 and 8.54. Mixing of the treated and untreated leachates maintained the pH value at near optimal conditions for nitrification without the need for pH control. When saturated with carbon dioxide after recirculation the leachate may return to lower pH levels but this may be buffered by the denitrification process, which increases the pH. The success of the treatment system may be dependent on the saturation of leachate with carbon dioxide and the anaerobic decomposition processes within the landfill.

The leachate within the treatment tank did not freeze during the winter of 1999 and treatment was initiated rapidly in 2000 with a relatively short (14 day) lag period. No further inoculation of treated leachate was required. It may have been possible to operate the system during the winter of 1999 but at a reduced rate to account for the drop in temperature which would have conflicted with the operation of the test cells prior to the commencement of the project.

There was no foaming associated with the treatment of leachate. The low organic content of the test cell leachate, typical of methanogenic leachates, and the general absence of volatile fatty acids, probably meant that there was less heterotrophic growth during treatment and less foaming associated with organic substances in the leachate. Precipitation of iron was evident on all the surfaces but biomass collection at the bottom of the tank was relatively limited. Sludge levels during settlement did not reach the outlet pipe and were held within the system. The sludge levels decreased over the winter months the tank was not operational to less than 1% v/v of the operational volume and nitrate levels decreased slightly over the winter of 1999-2000. Sludge levels in 2000 were approximately half that of 1999.

Settlement of the leachate prior to recirculation was 1 hour during which much of the bacterial biomass sank to the bottom of the aeration tank. Some finer material in the form of single bacteria cells were undoubtedly recirculated back into the landfill following emptying. The presence of biomedia and the surface area it offered for bacterial attachment certainly increased the performance of the aeration tank in nitrifying and oxidising the leachate. Much of the active biomass was attached to the biomedia where it persisted, ready for the next cycle. It was expected that the treatment during the first year would not be as successful as the second since the biomass

had to initially grow and become attached, to avoid flush out during recirculation. The presence of biomedia probably maintained bacterial activity rather than relying on suspended biomass.

5.2 Gas production from pulverised waste

Test Cell 3 contained pulverised refuse which was used previously during the Nitrogen Balances in Landfills project (Burton and Watson-Craik, 1997) and other laboratory studies on the decomposition of landfilled wastes. During one study on the ammonia fluxes during decomposition of refuse a 1 l system was constructed to accurately measure gas production by displacement (Burton and Watson-Craik, 1997). This system quantified the volume of gas released during decomposition by refuse at optimal conditions at 95% moisture content and possibly represented the maximum volume of gas that can be expected from the refuse. Experiments were performed in duplicate with landfilled refuse 1-2 month old landfilled pulverised refuse without a chalk buffer, which if used would have contributed to the total gas and the carbon dioxide production. The age of the landfilled waste used for this study was also appropriate in that it was similar to the age of the waste prior to capping and therefore it is appropriate to include it here for comparison. Larger plastics and metals were removed during preparation. Laboratory studies with 1-2 month old landfilled pulverised MSW produced 2.5 l gas from 53.2g dry weight equivalent to 47 m^3 $tonne^{-1}$ dry weight. Total methane production was equivalent to 28.2 m^3 $tonne^{-1}$ dry weight (Burton & Watson-Craik, 1997).

Gas flow data from the test cells during 1998 was incomplete. There were also omissions during 1999 and 2000 when the system was out of commission. Using very approximate measurements and a line of best fit (see Figure 4.8, $R^2 = 4.2$), the test cell produced approximately 5 m^3 h^{-1} (125 m^3 day^{-1}) in November 2000 which increased from the installation of the gas flow meters over a period of approximately 5 years. Calculating the 113875 m^3 for 3800 tonnes fresh weight (1330 dry tonnes) of emplaced refuse at approximately 65% moisture content, gives 30 m^3 gas $tonne^{-1}$ or 86 m^3 landfill gas $tonne^{-1}$ dry weight. The test cell thus greatly exceeded our laboratory experiments. In just under 5 years the Brogborough test cells produced between 30 and 50 m^3 gas $tonne^{-1}$ (Knox, 1995) for each of the different treatments. The Auchencarroch Test Cell compares well with these studies. Potential gas for the Brogborough cells was estimated to be approximately 200 m^3 $tonne^{-1}$. Test Cell 3 at Auchencarroch looks set to attain similar or greater gas production based on the observation that the gas flow rates show no evidence of decreasing.

From November 1998 to November 2000 the test cell was releasing between 3 - 4 m^3 h^{-1} (72-96 m^3 day^{-1}, 26208-35040 m^3 y^{-1}) which equates to 6.90-9.22 m^3 $tonne^{-1}$. From November 1999 to November 2000 the test cell was releasing between 4 - 5 m^3 h^{-1} (96-120 m^3 day^{-1}, 35040-43800 m^3 y^{-1}) which equates to 9.22 to 11.5 m^3 $tonne^{-1}$. Gas production was increasing despite a reduction in landfill temperature. These low temperatures will, as with all biochemical reactions, reduce microbial activity and methanogenesis.

The pulverisation of the waste undoubtedly increased the rate of gas formation allowing enhanced hydrolysis and bacterial activity as the cellulose fibres within the paper waste were more available to enzymic attack. The moisture content of the waste was also increased during the pulverisation process. The rapid capping that the test cell underwent compared to a normal landfill site where the landfilling operation can be prolonged may have also have been important.

The benefits of shredding and pulverising are by no means conclusive, and some authors describe a 'vigorous acid phase' that must be controlled if methanogenesis is not to be inhibited (Christensen *et al.*, 1992). Shredding or pulverisation introduce air into the refuse which may lead to high carbon dioxide concentrations in the pore spaces as the oxygen is consumed during respiration by aerobic and facultatively anaerobic bacteria. The Dano drum pulverisation process used introduced *extra water to help in the pulverisation (*pre-landfill tax) which may have reduced the penetration of air into the refuse following emplacement, as some of the paper waste was mixed into small lumps of papier-mâché.

5.3 Effects on the test cell following the recirculation of treated leachate

Nitrate introduced into the landfill was believed to be reduced to nitrogen gas by the process of denitrification. There is no direct evidence for this, as significant volumes of nitrogen gas could not be identified within the landfill. However, nitrate was removed from the leachate in the sump and net ammonia concentrations in the leachate decreased. The total volumes of nitrogen gas from the 14.7 kg NO_3^--N treated and recirculated during the experiment will have been negligible when compared to the landfill gas emitted by the landfill. A tracer experiment containing $^{15}NO_3^-$ would be required to identify $^{15}N_2$ gas above the trace amounts found in the gas vent. There was no evidence of a pattern in the gas concentrations in the days following the emptying of the aeration tank. Methane and carbon dioxide concentrations and the total gas analysed was similar throughout this period. Hydrogen gas was present only in trace amounts.

The depth of the cell from the head of the well did not vary significantly during the study. Following capping, little subsidence was recorded during the Field Trials in Waste Techniques project (EnviroCentre Ltd., 1997). The moisture content of the emplaced refuse was a little over 65% moisture content. The field capacity of the emplaced waste was believed to be slightly lower. We routinely extracted identical refuse from the source landfill site with moisture contents of between 50-65%. There was estimated to be approximately 2500 m^3 liquid in the test cell at the start of the project. The volume that could be treated and recirculated was estimated to be approximately 50 m^3 based on the recorded drop in leachate level of 0.6m over 3 month period following the first filling of the tank in 1999. A more accurate assessment of the volume in the saturated zone may be forthcoming in the final Phase II report of the Field Trials of Waste Manipulation Techniques project from Cell 1 following tracer studies and the removal of leachate from this test cell. A similar volume or head of leachate was aerated during the study since a total of 42 m^3 was recirculated back into the landfill site. A further 17.5 m^3 was removed and taken off site (see above) and 200 l added as seed.

Ammonia is released during the decomposition processes as microbial cells degrade and metabolise proteins in food and organic waste. The ammonia is removed by the deamination of the amino group forming free ammonia, which then equilibrates to ammonium leading to an increase in pH. Nearly all the ammonia will be present in solution as NH_4^+ at the pH values observed in the test cell and aeration tank. Release of ammonia in this way is often called ammonification. There were both saturated and unsaturated zones within the test cell used for this experiment. Ammonia will be present in solution and bound within the refuse. Refuse particles may enclose ammonia laden microsites which at moisture contents of 50% may never flush or equilibrate with the leachate for centuries. The ammonia will be bound to the refuse particles as

ammonium (NH_4^+). It may also be bound to the refuse but still have direct contact with the saturated zone and recirculating leachate. Ammonia may also be found in solution in pockets within the refuse particles. It is the binding and the interchange of ammonium and metal cations on these surfaces, which controls the ammonia content in the leachate. Therefore treating volumes of leachate equivalent to the saturated zone and returning it to the landfill will not see the removal of all ammonia in the leachate. Interchange promotes the return of ammonia into solution and its removal from bound surfaces. The interchange of leachate containing ammonia between the saturated and unsaturated zone will also allow ammonia to enter the leachate in the sump. There was approximately 2500m^3 of liquid in the site and just 50m^3 (1/50th of the volume) available for recirculation. Many other dissolved chemicals do not share this binding and once they are dissolved in the leachate interactions with the waste are probably infrequent. They thus remain in the leachate at constant levels once dissolved. Chloride and sodium dissolve easily into the leachate at high levels where they contribute significantly to the high conductivity of the leachate. They therefore represent better candidates for flushing from the refuse than ammonia.

Many of the refuse particles will possess a net negative charge in a similar way to soil particles and this promotes ammonia binding. Soil humus can be a good binder of ammonia, leading to concentrations which can only be released upon shaking and breaking up the soil fragments within a solution such as potassium chloride. These properties will make the refuse within the landfill capable of binding ammonia such that it can not be flushed from the landfill by perfusion or recirculation. Refuse columns perfused with distilled water during the previous Nitrogen Balances in Landfills project were flushed with over 200 bed volumes at a rate of 1 bed volume day^{-1} but the total nitrogen and the extractable ammonia concentration were virtually unchanged (Burton & Watson-Craik, 1997). Ammonia could not be analysed after 10 bed volumes. Breaking up the refuse in batch culture experiments at 80% moisture content increased the yield of ammonia in solution to between 5 and 10% of the total nitrogen content, close to that of the ammonia trapped within the columns after 200 bed volumes flushing with distilled water. Thus it is possible to characterise the ammonia in landfill site into a number of categories. Flushable ammonia represents the ammonia removed after approximately 10 bed volumes have been perfused through it. This is a truly arbitrary amount based on the techniques used during perfusion or recirculation of leachate. Extractable ammonia represents the true latent ammonia content based on an extraction by either shaking and blending in a solution of potassium chloride or using rapid distillation techniques. Total nitrogen content represents the ammonia in solution and within the protein in the refuse and in the microbial flora which are degrading it.

The ammonia concentration on 2.12.98 was 433 mg $NH_{3/4}^+$-N l^{-1} and similar concentrations were seen during most of the Field Trials of Waste Manipulation Techniques project. The approximate volume of leachate that could be recirculated was 50m^3 and contained an initial ammonia content of 21.6 kg $NH_{3/4}^+$-N. The amount treated and returned as nitrate was 14.7 kg $NH_{3/4}^+$-N with a further 6 kg removed from the landfill site either as nitrate or ammonia by tanker or transfer. Assuming that nitrate from the treatment was removed by the process of denitrification in the landfill, sufficient ammonia was exchanged with the refuse to keep the ammonia concentration at 290 mg $NH_{3/4}^+$-N l^{-1} on 6.2.01. Ammonia probably entered the leachate through interchange with the moisture within the refuse and by interaction with ammonia bound on the refuse particles.

The pulverised refuse from Shewalton used in Cell 3 during previous laboratory perfusion studies was analysed and contained 1.59 (SD 0.210) % nitrogen dry weight (Burton & Watson-Craik,

1997). From these perfusion studies, only 4.41 (SD 0.547) % of the total was leachable within 50 bed volumes (Burton & Watson-Craik, 1997). This was similar to the percentage that could be extracted from batch culture studies at 80% moisture content (Burton & Watson-Craik, 1996a). Using the data from the perfusion studies, approximately 969 kg ammonia in the test cell was leachable of which 20.7 kg was removed or treated. Thus only 2.13% of the extractable ammonia would have been removed from the test cell, assuming that the recirculated nitrate was consumed by the denitrification process. It would have been possible to treat much more ammonia from the test cell with a much cheaper lagoon system in a matter of months, but this would have interfered with the routine recirculation of 7 to 7.5 m^3 leachate. However, the employed treatment system introduced significant concentrations of nitrate into the landfill when it was emptied. These concentrations of nitrate did not appear to significantly inhibit methanogenesis within the test cell.

5.4 Future work

The Auchencarroch Test Cells have yielded a considerable amount of data over a relatively long time period. Gas flow, temperature and gas concentrations have been monitored. Much is known about the waste that went into the test cells and the nitrogen balances occurring during the decomposition of the pulverised refuse used in Cell 3 and Cell 1. The pulverised refuse has also been subjected to permeability studies using a specially constructed compression cell (Hudson *et al.*, 2000). These research programmes funded by the EA will soon be paying dividends, as there will be an opportunity to collate much of the information into a landfill bioreactor model.

6. CONCLUSIONS

Leachate could be successfully treated in an aeration tank during experiments on the acceleration of the decomposition of pulverised refuse in a landfill test cell. Treated leachate containing nitrate was recirculated, returning it to the landfill test cell. Routine analysis of the test cell leachate did not reveal major differences in the ammonia concentrations by the end of 1999, but by 2001, ammonia concentration were significantly lower despite recirculation without treatment. Gas production was not inhibited. There was evidence that gas production increased during the experiment. Increases in gas flow could not be attributed directly to recirculation of nitrified leachate.

A strategy of recirculating treated leachate can be used to accelerate landfill decomposition and landfill attenuation by reducing ammonia concentrations. Cost remains a significant hurdle in suggesting this strategy as a method of reducing ammonia concentrations. In previous reports available in the CWM series we have suggested that filter beds of crushed rock above the cap could be used which would enable some in-situ treatment prior to recirculation. If this system could be constructed without affecting the integrity of the landfill then this may offer a much cheaper solution. The recirculation and treatment system described in this report was constructed as a closed system and was therefore designed to operate within the landfill recirculation regime carried out on site prior to installation. The recirculation of nitrified leachate can offer a method of reducing ammonia concentration without affecting gas production. In light of the EC Landfill Directive (1999/31EC) it is unlikely that a full-scale landfill bioreactor will ever be commissioned in the UK and the strategy of recirculating nitrified leachate applied to it. The strategy however may find an application in currently operated landfill sites where in combination with an effective recirculation system, it can reduce inhibitory concentrations of ammonia and volatile fatty acids in the landfill through treatment and dilution.

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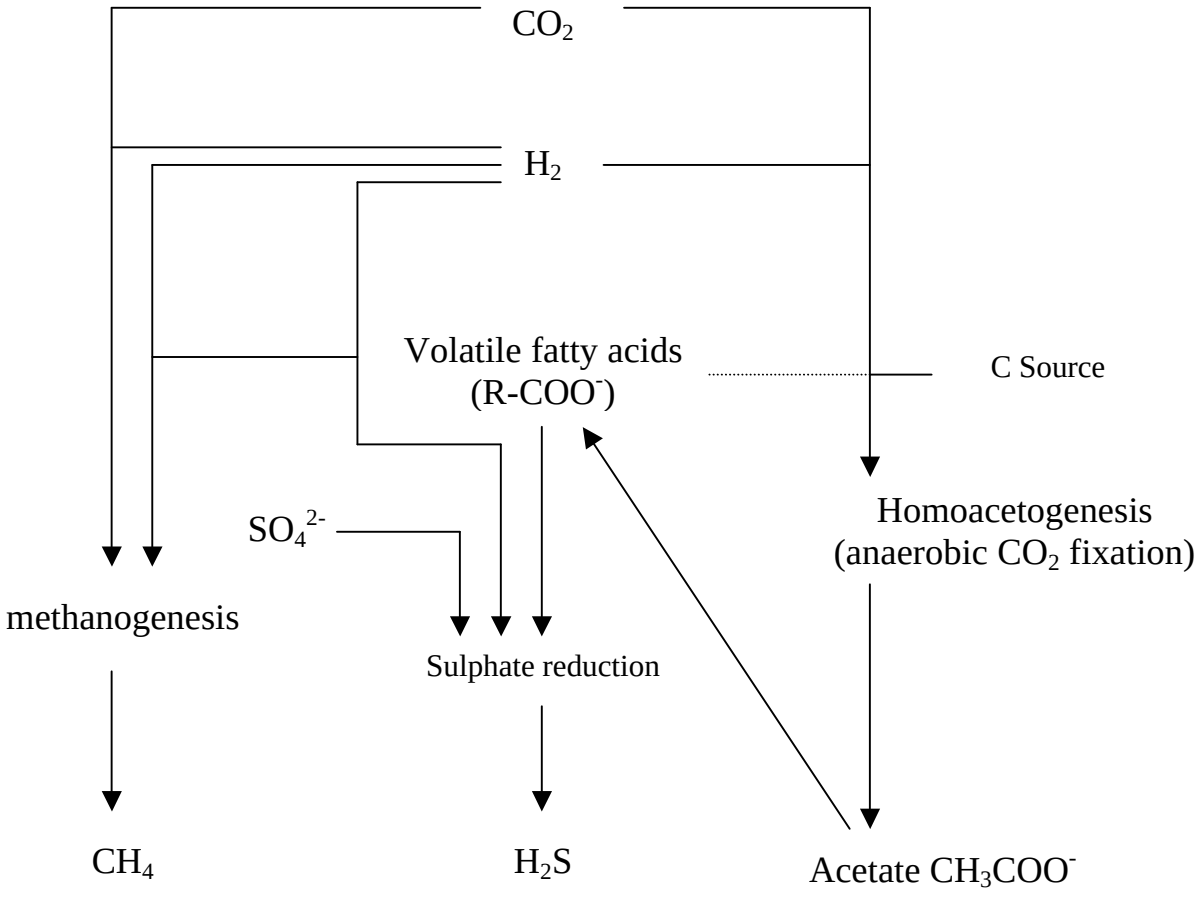
Appendix 1: Refuse Decomposition Processes

Paper, card, and vegetable and animal mater represent the chief biodegradable portion of refuse. The decomposition of refuse is usually anaerobic in nature and carried out by bacteria in the absence of air. Moisture promotes the solubilisation of the wastes and facilitates bacterial decomposition through enzymic attack. The chief decomposition products are detailed below:

Paper (Cellulose) ?
(1) landfill gas methane CH_4 (60%) and carbon dioxide CO_2 (40%) and trace gases and odours such as hydrogen sulphide H_2S.
(2) Mixture of carboxylic acids
(3) Hydrogen H_2 (trace)

The decomposition of waste is complicated and involves different functional groups of bacteria which routinely transfer substrates during energy production and growth. The interdependency of this relationship contributes to lags in methane production and can lead to high concentrations of volatile fatty acids prior to the onset of methane production. Different functional groups of bacteria such as acetogens which release acetate, homoacetogens, methanogens and sulphate reducing bacteria each have an important roles in controlling overall refuse decomposition. The relationship between the tertiary bacterial processes of refuse decomposition and the key products of the anaerobic decomposition of paper waste, CO_2, H_2, volatile fatty acids and methane are shown below:



Appendix 2: Analytical data from test cell 3

Appendix 2 contains the routine analytical data from Test Cell 3 at the DoE and EA funded Auchencarroch Test Cell Facility. It shows the routine analysis of the leachate from the test cell at the start of the Field Trials of Waste Manipulation Techniques project, on 22.11.95, to the last analysed sample on 6.2.01. The final recorded entry for the Field Trials of Waste Manipulation Techniques project displayed was on 20.8.97 (EnviroCentre Ltd., 1998). The Ammonia Fluxes in Landfills Project continued until the winter of 2000. During 1998 the samples from the test cells were removed and sent for analysis by EnviroCentre Ltd. (data not shown). Samples were also at the same time removed by the authors and frozen for reference. The authors, prior to freezing, performed measurements of pH and conductivity on these samples. Samples were also analysed for nitrate and volatile fatty acids.

The frozen samples were defrosted over two days in the authors' laboratory and then collected by the Caledonian Shanks Centre for Waste Management, Glasgow Caledonian University for analysis by Bodycote Ltd. One sample, taken on 2.12.98, was sent by the authors unfrozen for analysis by Bodycote Ltd., and then sent again with the other frozen samples from 1998. This sample acted as a control for the freezing and defrosting process and is shown in red. Frozen samples are displayed in blue. During 1998 the maintenance contract for the Field Trials of Waste Manipulation Techniques project was transferred to the Caledonian Shanks Centre for Waste Management, Glasgow Caledonian University. With the exceptions stated above, the majority of the data presented from 1999 onwards was processed by the Caledonian Shanks Centre for Waste Management, Glasgow Caledonian University and analysed by the subcontractor Bodycote Ltd.

The sample data displayed were the product of a single analysis and were not performed in duplicate or triplicate like the other analyses performed in the authors' laboratory during the Ammonia Fluxes in Landfills project. The data will be republished and updated in the final Phase II report of the Field Trials of Waste Manipulation Techniques project by the Caledonian Shanks Centre for Waste Management, Glasgow Caledonian University. The data were received from the Caledonian Shanks Centre for Waste Management, Glasgow Caledonian University on 6.11.00 and 7.3.01. The sampling of Cell 3 during the Ammonia Fluxes in Landfills project was usually carried out by the authors or with the assistance of the holders of the site maintenance contract.

Date	Acetic acid	Alkalinity	Ammoniacal nitrogen	Biochemical oxygen demand	Cadmium	Calcium	Chemical oxygen demand	Chloride	Chromium	Copper	Electrical Conductivity
22/11/95	2895.00	5737	613	8500	0.03	1499	13720	1055	0.20	0.48	11900
20/12/95		4435	432	4000		145	6950	1295			10600
18/01/96		3480	373	600		70	1970	1205			9095
21/02/96	17.00	716	161	200		58	1800	1205			9220
20/03/96		4225	406	675		59	1750	1330			10400
18/04/96		3930	415	100		35	1610	1240			10250
21/05/96	24.50	3660	358	135		311	1400	1310			9705
27/06/96		3775	348	45		381	1200	1225			9890
17/07/96		3840	392	85		356	1280	1225			9875
21/08/96	10.00	3940	370	30		347	1050	1195	0.08	0.07	9840
05/09/96		3885	255	200		466	1450	1195			9775
20/10/96		3645	255	95		433	1325	1160			9680
20/11/96		3890	338	180	0.02	477	1425	1210	0.17	0.01	7840
20/12/96		4010	351	200		360	1450	1260			9235
20/01/97		3850	295	80		318	735	1200			8555
20/02/97	12.00	3795	355	93		506	1105	1245			6700
20/03/97		4085	339	95		353	1750	1255			9495
20/04/97		4000	833	155		450	1095	1210			8455
20/05/97		3810	406	95		372	1385	1270			8585
20/06/97		4145	394	130		342	1325	1265			8845
20/07/97		4120	423	3150		295	1315	980			2555
20/08/97		3825	360	15		308	1455	1260			8800
22/10/97											
05/11/97											
19/11/97											9680
23/01/98			411			1	913	245			
04/02/98											9970

Date	Acetic acid	Alkalinity	Ammoniacal nitrogen	Biochemical oxygen demand	Cadmium	Calcium	Chemical oxygen demand	Chloride	Chromium	Copper	Electrical Conductivity
18/03/98			408			7	888	261			9880
18/04/98											9780
20/05/98			428			16	915	259			9750
17/06/98											9930
15/07/98			422			9	893	231			9940
29/07/98											9910
12/08/98			444			23	853	218			9910
07/10/98											9660
04/11/98											9410
02/12/98			425			31	748	286			
02/12/98	<5	3472	433	85	0.01	86	773	1000	0.07		8538
02/03/99		3540	420	78		63	746	941			8059
23/04/99	<5	3466	224	331		223	1229	707			9300
05/05/99	<5	3460	396	115		295	768	912			9303
08/06/99		2791	297	76		197	630	659			7488
22/06/99		1225	113	76		100	258	233			2531
06/07/99		1960	241	24		112	438	376			6006
21/07/99		2022	223	211		2	308				5308
03/08/99		4023	441	46		65	788	1040			10207
17/08/99		4697		35		93	2675	846			12796
03/09/99		3431	368	<3		111	788	732			9303
14/09/99		3247		82		21	708	811			867
01/10/99		3410	348	<3		1301	805	761			
12/10/99		817	71	10		1	388	145			1776
29/10/99		1016	398	<3		250	793	992			8059
10/11/99		4058	411	97		210	865	960			11020

Date	Acetic acid	Alkalinity	Ammoniacal nitrogen	Biochemical oxygen demand	Cadmium	Calcium	Chemical oxygen demand	Chloride	Chromium	Copper	Electrical Conductivity
23/11/99		12173	246	48		434	750	862			12173
23/12/99		3789	403	124		263	883	1057			10789
11/01/00		3549	365	70		105	778	1081			10965
26/01/00		3823	395	52	<0.005	107	805	1021	<0.005	<0.01	8033
22/02/00		3708	392	8		295	783	925			8843
07/03/00		3689	380	10		308	703	972			8741
04/04/00		3752	386	67		50	793	1236			8843
18/04/00		3335	366	38		202	865	936			9116
02/05/00		3842	324	19		187	800	851			8843
16/05/00		3539		41		115	482	823			7832
30/05/00		3235	332	62		37	464	767			8979
14/06/00		3943	405	30.70		12	1195	855			9841
28/06/00		3084	415	90.00		15	977	790			9841
25/07/00		3918	411	61.00		0.20	882	1024			9391
09/08/00		2629	130	22.10		2.00	377	514			6844
05/09/00		2376		46.10		0.40		610			4136
19/09/00		556	90	118		85.00	414	230			2097
06/10/00		3185	171			1.20					5561
31/10/00		3478	302	47		186.00	766	1364			7899
17/11/00		3448	282	54	<0.005	222.00	643	681	0.02	0.03	7766
30/11/00		3286	342	142		89.00	614	871			7500
12/12/00		3033	259	33	0.02	461.00	614	1066	0.04	0.03	7567
23/01/01		3286	311	59		155.00	861	891			7766
06/02/01		3437	290	115		179.00	680	937			8707

Date	Iron	Iso-butyric acid	Iso-valeric acid	Lead	Magnesium	Manganese	N-butyric acid	N-valeric acid	Nickel	Nitrate	Nitrite
22/11/95	210.00	465.00	415.00	0.10	264.0	26.00	2280.00	930.00	0.05	36.00	0.10
20/12/95					200.0					10.25	0.10
18/01/96					174.5					5.15	0.10
21/02/96	1.53	10.00	10.00		176.0	0.02	10.00	10.00		4.50	0.10
20/03/96					152.0					3.90	3.90
18/04/96					157.5					4.75	0.10
21/05/96	16.60	10.00	10.00		186.0	1.14	10.00	10.00		5.00	0.10
27/06/96					193.5					1.00	0.10
17/07/96					193.5					1.00	0.10
21/08/96	24.35			0.08	184.0	1.08			0.22	1.00	0.10
05/09/96					197.5					1.00	0.10
20/10/96					199.0					1.00	0.10
20/11/96	17.45			0.03	215.0	1.41			0.29	1.00	0.10
20/12/96					177.5					1.00	0.10
20/01/97					183.5					1.00	0.10
20/02/97	24.60	10.00	10.00		252.5	1.23	10.00	10.00		1.00	0.10
20/03/97					225.0					1.00	0.10
20/04/97					291.5					1.00	0.10
20/05/97	20.80				239.0	1.00				1.00	0.10
20/06/97					224.5					1.00	0.10
20/07/97					205.0					2.55	0.10
20/08/97					205.5					1.00	0.10
22/10/97											
05/11/97											
19/11/97											
23/01/98					52.0					<1	<1
04/02/98											

Date	Iron	Iso-butyric acid	Iso-valeric acid	Lead	Magnesium	Manganese	N-butyric acid	N-valeric acid	Nickel	Nitrate	Nitrite
18/03/98					152.0					<1	<1
18/04/98											
20/05/98					216.0					<1	<1
17/06/98											
15/07/98					137.0					<1	<1
29/07/98											
12/08/98					232.0					<1	<1
09/09/98											
07/10/98											
02/12/98					173.0					<1	<1
02/12/98	2.50	<5	<5		157.0	0.13	<5	<20		<1	<1
02/03/99					64.0					1.10	2.50
23/04/99	21.00	<5	<5		89.0	1.10	<5	<5		<1	<1
05/05/99	28.00	<5	<5		119.0	1.10	<5	<5		<1	<1
08/06/99					111.0					<1	<1
22/06/99					45.0					<1	1.20
06/07/99					192.0					<1	<1
21/07/99					1.0						
03/08/99					293.0					<1	2.50
17/08/99					190.0					<1	<1
03/09/99					266.0					<1	<1
14/09/99					172.0					<1	<1
01/10/99					1239.0					<1	<1
12/10/99	<0.1				3.3	0.28				<1	<1
29/10/99					121.0					<1	<1
10/11/99					219.0					<1	<1
23/11/99	19.00				110.0	1.80				<1	<1

Date	Iron	Iso-butyric acid	Iso-valeric acid	Lead	Magnesium	Manganese	N-butyric acid	N-valeric acid	Nickel	Nitrate	Nitrite
23/12/99					58.0					<1	<1
11/01/00	8.50				85.0	2.40				<1	<1
26/01/00	5.00			<0.05	113.0	0.48			0.23	<1	<1
22/02/00					136.0					<1	<1
07/03/00					198.0					<1	<1
04/04/00					301.0					<1	<1
18/04/00	5.90				153.0	0.01				<1	<1
02/05/00					134.0					<1	<1
16/05/00	0.30				10.0	0.01				<1	<1
30/05/00	25.00				196.0	2.46				<1	<1
14/06/00					0.6					<1	<1
28/06/00					0.1					<1	<1
25/07/00	0.20				97.0	0.01				<1	<1
09/08/00	0.10				1.0	0.02				<1	<1
05/09/00	0.49				0.5	0.04				<1	<1
19/09/00					37.0					<1	<1
06/10/00					20.0						
31/10/00					109.0					<1	<1
17/11/00	0.30			<0.05	139.0	<0.01			0.04	<1	<1
30/11/00					80.0					<1	<1
12/12/00	28.00			<0.05	381.0	0.80			0.24	<1	<1
23/01/01					128.0					<1	<1
06/02/01					132.0					<0.1	<0.1

Date	Phosphate	pH	Potassium	Propionic acid	Sodium	Sulphate	Temperature	Total organic carbon	Total oxidised nitrogen	Total volatile acid	Zinc	Total Kjeldahl N
22/11/95	0.20	6.80	256.50	2700	864	164	28.20	6850	36.00	9690	0.95	
20/12/95		7.00	342.5		919		22.90	2450				
18/01/96		7.45	275.0		820	61	16.10	816				
21/02/96	0.55	7.10	277.0	10	923	49	21.65	490	4.45	34		
20/03/96		7.10	426.5		1100	43	27.20	315				
18/04/96		7.00	365.5		1007	119	24.00	467				
21/05/96	0.95	7.05	363.0	10	1015	48	24.30	405	5.00	31		
27/06/96		7.00	371.0		1140	49	22.00	455				
17/07/96		6.50	357.5		1035	48	20.70	299				
21/08/96	0.95	7.10	401.0		1100	43	16.40	345	1.10	21	0.30	
05/09/96		7.10	302.5		943	50		350				
20/10/96		7.00	341.0		988	46		316				
20/11/96		7.20	308.0		1025	45		396	329.50		0.25	
20/12/96		7.00	336.5		1130	38	16.90	375				
20/01/97		7.00	176.0		578	42	14.30	185				
20/02/97		7.00	282.5	10	1129	53	13.20	330	406.50	24		
20/03/97		7.00	382.5		1315		15.90	200				
20/04/97		7.10	372.0		1260	46.90	15.40	360				
20/05/97	0.40	7.20	324.0		1055	32.30	14.60	335	408.50	109		
20/06/97		7.30	348.0		1185	40.40	16.10	395				
20/07/97		7.20	300.0		1100	0.50	14.80	1321				
20/08/97		7.20	337.5		988	39.30	17.10	470				
22/10/97		6.99										
05/11/97		6.94										
19/11/97		7.02										
23/01/98	0.20	7.01	180.0		233	14						605.00
04/02/98		7.00										
18/03/98	0.60	6.92	319.0		597	17						594.00

Date	Phosphate	pH	Potassium	Propionic acid	Sodium	Sulphate	Temperature	Total organic carbon	Total oxidised nitrogen	Total volatile acid	Zinc	Total Kjeldahl N
18/04/98		6.94										
20/05/98	2.60	6.88	373.0		843	14						580.00
17/06/98		6.88										
15/07/98	0.60	6.90	350.0		550	17						580.00
29/07/98		7.00										
12/08/98	2.30	6.92	447.0		966	14						568.00
09/09/98		7.02										
07/10/98		6.90										
04/11/98		6.99										
02/12/98	2.10		449.0		659	24						540.00
02/12/98	0.12	7.60		<5	1172	58		299	<1	<5		
02/03/99		7.00	177.0		581	3.10	20.00	246				
23/04/99	1.80	7.10	139.0	<5	605	<1	25.00	335		<5		323.00
05/05/99	2.10	6.90	163.0	<5	596	3.80	25.00	190		<5		296.00
08/06/99		6.90	276.0		478	<1		211				
22/06/99		6.70	166.0		276	1.20		116				
06/07/99		7.10	212.0		554	<1	20.00	161				
21/07/99		7.00	265.0		13			312		<5		
03/08/99		6.70	269.0		1610	<1	22.00	286				
17/08/99		7.30	469.0		895	<1	22.00	529				
03/09/99		7.00	307.0		143	<1		430				
14/09/99		6.90	368.0		723	<1		158				
01/10/99		6.90	251.0		4978	<1		211				
12/10/99	0.20	6.30	56.0		17	<1		93		<10		455.00
29/10/99		6.90	164.0		1295	<1		244				
10/11/99		7.20	342.0		1225	60.00		252				
23/11/99	2.70	7.10	472.0		1391	<1		242		<10		
23/12/99		6.90	211.0		847	<1		214				

Date	Phosphate	pH	Potassium	Propionic acid	Sodium	Sulphate	Temperature	Total organic carbon	Total oxodised nitrogen	Total volatile acid	Zinc	Total Kjeldahl N
11/01/00	5.40	6.90	345.0		533	<1		235		<15		687.00
26/01/00	1.40	7.20	279.0		775	<1		226		<15	0.12	492.00
22/02/00		7.20	874.0		1276	<1		28				
07/03/00		7.00	294.0		1625	2025.00		16				
04/04/00		7.00	353.0		1623	<1		250				
18/04/00	8.60	7.00	343.0		775	<1		320		<10		438.00
02/05/00		7.00	303.0		1447	13		207				
16/05/00	10.00	7.50	2756.0		406	76		170	<1	<1		460.00
30/05/00	1.80	7.90	2978.0		1265	87		396	<1	<1		420.00
14/06/00		7.10	1.4		127	144		785				
28/06/00		7.10	215.0		645	111		7				
25/07/00		7.10	2.1		11	129		390				418.00
09/08/00	0.45	7.10	1.0		1	91		198				287.00
05/09/00	3.40	6.80	1.6		15	68		103				258.00
19/09/00		6.20	69.0		1548	<1		102				
06/10/00			2755.0		384							
17/11/00	6.60	7.00	227.0		588	17.00		927		<1	<0.01	670.00
30/11/00		6.90	130.0		380	22.00		171				
12/12/00	31.00	7.20	551.0		1780	3.90		149		<1	0.05	444.00
23/01/01		7.00	196.0		683	54.00		1055				
06/02/01		7.00	216.0		1029	55.00		215				