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REMOVAL OF AMMONIACAL NITROGEN FROM LANDFILL LEACHATE BY IRRIGATION ONTO VEGETATED TREATMENT PLANES

S.F. TYRREL, P.B. LEEDS-HARRISON, & K.S. HARRISON

Institute of Water and Environment, Cranfield University, Silsoe, Bedford, MK45 4DT, UK

Tel: 44 (0) 1525 863293 Fax: 44 (0) 1525 863344 s.tyrrel@cranfield.ac.uk

ABSTRACT

Leachate is a contaminated liquor resulting from the disposal of solid and liquid wastes at landfill sites that must be treated before discharge. Vegetated leachate treatment planes have been used at landfill sites in the UK but have received little scientific attention. This paper describes studies of model leachate treatment planes with a focus on the removal of ammoniacal nitrogen (NH_3-N). Small-scale and field-scale experimental treatment planes were constructed, filled with clay loam soil and vegetated with grass *(Agrostis stolonifera)*. Landfill leachate was applied at hydraulic loading rates ranging from 17-217 l/m^2/d. An exponential relationship was used to characterise the pattern of NH_3-N removal. No relationship was observed between the hydraulic loading rate and the NH_3-N removal rate constants (R^2 = 0.0039). The daily specific NH_3 -N mass removal rate was found to be linearly related to the NH_3 -N concentration at the start of that day of treatment (R^2 = 0.35). Possible causes of variation in the rate of NH_3-N removal between experiments are discussed. A simple inorganic nitrogen balance indicated that the mass of NH_3-N and NO_2-N removed was not accounted for by NO_3-N production. Explanations for this apparent nitrogen deficit are discussed.

Key words – landfill, leachate, ammonia, wastewater treatment

INTRODUCTION

Landfilling is the disposal of waste in void spaces created by activities such as gravel or clay extraction. The practice of landfilling results in a number of potentially damaging environmental impacts of which the generation of landfill leachate is just one. In addition to the initial moisture content of the solid waste and any liquid waste inputs, water may enter landfills as a result of the ingress of precipitation, surface water or groundwater. Contact between this water and the waste generates a leachate contaminated with a range of soluble organic and inorganic substances. Generally, leachate generated from recently-filled domestic waste has a high 5 day biochemical oxygen demand (BOD_5), of about 10 000 mg/l, and a total ammoniacal nitrogen (NH_3-N) concentration in excess of 1000 mg/l. This is a result of the microbiological degradation of organic substances under anaerobic conditions (Robinson and Maris, 1979; Robinson and Gronow, 1992; Robinson, 1995). In older landfills the readily biodegradeable compounds contributing to BOD_5 are converted by a complex series of microbiological reactions to methane and carbon dioxide and may be extracted from the fill as landfill gas. As there is no significant mechanism for NH_3 -N transformation in the landfill, NH_3 -N concentrations remain high in aged wastes. Landfilled waste will continue to produce leachate contaminated with NH_3 -N for many years after filling operations have ceased (Robinson and Gronow, 1992).

Leachate in an untreated form is unsuitable for direct discharge into surface watercourses as the high BOD and NH_3-N concentrations would have a severe impact on the ecology of the receiving water. Although effective advanced leachate treatment systems exist, some landfill operators seek alternatives because of their high capital costs and specialised management requirements. Land-based treatment systems are an attractive alternative for landfill operators as they utilise an existing land resource, they are considered to be cheap to build and operate, and are not perceived to need sophisticated management. A land-based system is often seen as a “stop-gap” measure, permitting the investment in an expensive, advanced leachate treatment system to be deferred, perhaps until the time when filling operations cease. Alternatively, land-based systems may be used in conjunction with a conventional tank-based system to play a polishing role (Maehlum, 1995; Martin and Johnson, 1995).

An example of an extensive, land-based system adopted at landfill sites in the UK is the leachate treatment plane. Treatment planes are areas of vegetated, sloping land, usually constructed using low permeability soils onto which leachate is applied from a recirculation lagoon. Overland flow of leachate is encouraged by the slope, the lack of vertical percolation and the high rate of wastewater application. As a result, leachate interacts with the upper layers of the soil and the vegetation as it returns to the recirculation lagoon, hence the use of the generic term “overland flow” treatment system. The process of recirculation continues (perhaps for several weeks) until the entire batch of leachate in the lagoon reaches the required quality standard for discharge.

A limited amount of research has been undertaken into mechanisms of NH_3-N removal on overland flow treatment systems. The principal work was reported by Kruzic and Schroeder (1990) following studies of systems used for the treatment of settled sewage. In a series of experiments using single-pass, laboratory-scale treatment systems, they concluded that:

- a retention process (probably cation exchange) was responsible for NH_3-N removed one day and then being released in the form of nitrate on a subsequent day;

- nitrification is a significant process (high nitrate concentrations in the treated wastewater provided evidence of this);
- denitrification is responsible for the removal of some of the NO_3-N produced as a result of nitrification.

Although leachate treatment plane systems have been in use in the UK for many years, little is known of the treatment mechanisms involved or the factors affecting treatment efficiency. This paper reports the findings of studies of experimental treatment planes designed to improve our understanding of ammoniacal nitrogen dynamics in these engineered grassland treatment systems.

METHODOLOGY

Introduction

Operational treatment planes may have a surface area of several hectares so it was considered desirable to build an experimental system as large as practically possible. Accordingly, 10 separate 25 m long x 1 m wide field-scale plots were constructed at Silsoe, Bedfordshire, UK. In addition to the field-scale plots, further data were obtained from experiments carried out using 2 m long x 0.4 m wide small-scale troughs.

Field-scale plots: design, construction and operation

The first stage of construction was to excavate a pit (approximately 16 m long x 2.5 m wide x 2 m deep) to hold the 1.2 m^3 reception tanks which would collect leachate runoff from the plots. A reinforced concrete base was laid in the bottom of the pit and tied to reinforced, hollow-block retaining walls. Treatment plane preparation began on completion of the pit. 0.4 m of topsoil was removed, using an excavator, from a 25 m x 16 m area adjacent to the pit and stockpiled for later use. The excavator was then used to grade the site to give a 2% down-slope gradient and a 0% cross-slope gradient with the lower end of the slope meeting the edge of the pit retaining wall.

Ten plots were then marked out in five pairs, each pair separated by a 1 metre wide pathway for access (Figure 1). A timber frame was erected on each plot and, to prevent seepage from the plots, a polyethylene sheet was positioned over the wooden frame, covering each pair of plots as shown in Figure 2. The stockpiled topsoil clods were broken up to form smaller, more manageable aggregates with a power harrow and carefully replaced inside the polythene-lined plots. The soil was then irrigated with fresh water for 8 hours and allowed to consolidate for a week. High and low areas on the plot surfaces were evened out by raking and the level checked at 1 m intervals downslope to ensure that a uniform 2% gradient was achieved.

Seed of the grass *Agrostis stolonifera* was sown on the completed plots, at the supplier's recommended rate (50 g/m^2). *A. stolonifera* was selected because it was a dominant member of the grassland community on two local treatment planes. At both sites, the grass appeared to tolerate periodic inundation with leachate with no apparent ill effects. In pot trials, *A. stolonifera* has outperformed (Bradford, 1992; Boon, 1996; Elliot, 1997), other salt-tolerant and waterlogging-tolerant grass species (e.g. *Spartina townsendii* and *Pucinellia maritima*) when irrigated with raw leachate and can survive for weeks submerged in water containing high concentrations of chloride.

A 1.2 m^3 delivery tank was placed on the bund at the upper end of each of the plots. Landfill leachate for the experiments came from Shanks Ltd.'s Calvert landfill site in

Buckinghamshire, UK. Calvert opened in 1980 and is classified by Robinson (1995) as a large, relatively dry landfill with a high waste input rate. The leachate used in these experiments came from a part of the landfill characterised as methanogenic with a low BOD (typically < 200 mg/l). A water quality summary of the raw Calvert leachate during the experimental period is given in Table 1. Leachate from the delivery tanks was applied at the top of the plots using a simple weir arrangement. Runoff from the plots was collected in two linked 1.2 m^3 reception tanks at the base of the slope.

The plots were operated on a 24 hour cycle. Leachate application began at 10 *a.m*. and continued for 5 hours. The soil was then allowed to drain and dry for 19 hours until the next application period began the following day. In their work with overland flow systems for the treatment of domestic wastewater, Smith and Schroeder (1985) demonstrated that continuous (24 hr/d) application was deleterious to the treatment process. A drying period allows atmospheric oxygen to diffuse into the soil, permitting the processes of organic matter and NH_3 -N oxidation to proceed. The leachate was pumped back into the delivery tanks every morning at which point samples were taken for chemical analysis.

Small-scale troughs: design, construction and operation

2 m long x 0.4 m wide x 0.2 m deep troughs were constructed from mild steel sheet following a design used previously by Pawson (1993)(Figure 3). The troughs were partially filled to a depth of approximately 0.1 m with the clay loam soil (Evesham series) found at the experimental site at Silsoe. The soil was then covered with turf, taken from an operational treatment plane run by Shanks at Calvert in Buckinghamshire, UK. The predominant grass species present was *Agrostis stolonifera*

(creeping bent). The troughs were positioned such that they had a 2% down-slope gradient and a 0% cross-slope gradient. Leachate was applied to each trough from its own 220 l reservoir (a polypropylene barrel) that discharged its contents via a control valve to a transverse distributor pipe positioned at the upper end of the trough. At the lower end of the trough, leachate runoff was collected in an identical 220 l polypropylene barrel. The troughs were operated in the same ways as the field-scale plots.

Plot and trough experimental programme

The principal experimental variable under investigation was the hydraulic loading rate. The hydraulic loading rate has the potential to influence the functioning of the system in a number of ways. At low hydraulic loading rates leachate may not be well distributed leading to poor contact with the available surface area of the treatment plane. At higher hydraulic loading rates depth of flow will increase. This will in turn lead to higher velocities and lower residence times. Hydraulic loading rates were tested within the range 17 - 217 l/m^2/d (17-217 mm/d)(Table 2 & Table 3). This compares with estimated hydraulic loading rates of 52-72 l/m^2/d (52 mm/d-72mm/d) at treatment planes operated by Shanks Ltd.

A start up plot trial served to draw attention to practical problems associated with the larger-scale experiment. It was found that the 1.2 m^3 capacity of the delivery tanks was insufficient to sustain an experimental run of longer than 2 weeks on a 25 m^2 plot due to water loss by evapotranspiration. For example, during hot weather, evapotranspiration could be 3 mm/d, equivalent to 75 l per 25 m plot per day. This problem was overcome in two ways. Firstly, plot length (and therefore area) was reduced by moving the distributor trough down the plot. Thus plot lengths of 10 m or 20 m were used to increase the hydraulic loading rate. Secondly, the delivery tank volume of each plot was increased by linking the delivery tanks for two plots together to feed a single plot.

Sampling and analysis

Leachate samples were taken every weekday morning following the return of the leachate treated on the previous day from the reception tanks to the delivery tanks. Prior to sampling, the contents of each delivery tank were vigorously stirred to ensure that treated leachate and any untreated leachate were thoroughly mixed (at low hydraulic loading rates, not all the delivery tank volume was discharged each day). All leachate samples were immediately refrigerated and transported in a cool box for laboratory analysis. NH_3-N, NO_2-N and NO_3-N were measured daily by colorimetry using a Hydrocheck 600C spectrophotometer (WPA Ltd, Cambridge, UK). COD, Cl^-, and pH were also measured daily but for the sake of brevity, these data are not discussed.

RESULTS AND DISCUSSION

NH_3-N removal dynamics

The recirculation of leachate over experimental treatment planes leads to a slow, progressive reduction in its NH_3-N concentration over a period of several weeks. A similar pattern of NH_3-N removal was produced in both the trough and the plot experiments. NH_3-N removal was relatively rapid during the first few days but declined as the experiment proceeded. This generally gave the appearance of a curve when NH_3-N concentration was plotted against experimental days (Figure 4). An exponential relationship was found to best fit the experimental data. The best-fit line equations and the associated coefficients of determination (R^2) for each of the trough and plot experiments are presented in Table 4. The value of k (the reaction rate constant), derived from the best-fit line equations, was plotted against the corresponding hydraulic loading rate for each experimental run (Figure 5). The scattered points and the associated low value of R^2 (0.0039) suggest that there is no relationship between hydraulic loading rate and the rate of NH_3-N removal. This suggests that factors likely to be related to hydraulic loading rate, such as uniformity of leachate distribution and leachate residence time, were in fact insignificant in terms of their influence on treatment performance.

Nevertheless, the values of the NH_3-N removal rate constant for each of the experimental runs varied considerably. Land-based treatment systems are complex and a number of factors may influence the rate of NH_3-N removal during the course of an experiment. Spatial variation in the NH_3-N adsorption capacity and the state of the nitrifying bacteria populations of the soil used might be expected. At the lower end of the range of application rates used it is likely that contact between the treatment plane and the leachate would be diminished due to preferential flow. Temperature is also likely to have had an affect on the NH_3-N removal rate by affecting the rate of nitrification and, to a lesser extent, other microbiological and chemical processes. As the experiments extended from early spring through to early autumn, it is reasonable to expect that temperature would have contributed to the level of variation observed. The chemical and biological history of a given area of treatment plane is another factor that may have contributed to the variation observed. For example, it may take several days for nitrifying bacteria in the soil to build up to optimum treatment levels following the initiation of leachate application. Soil that is receiving leachate for the first time may differ microbiologically to soil that has received leachate many times before. Similarly, it is conceivable that chemicals may accumulate in the soil following repeated applications of leachate that may influence chemical adsorption or microbiological degradation processes. Differences in raw leachate chemistry could also affect the microbiological activity of the treatment plane and therefore contribute to differences in the NH_3-N removal rate between experimental runs. High nitrite concentrations, which were present in some of the batches of raw leachate taken from the Calvert landfill site, could for example have had a negative effect on NH_3-N oxidising bacteria through end product inhibition. Whilst it is reasonable to expect that all of the above mentioned factors could have contributed to the variation observed, the relative significance of individual factors is unknown.

A generalised NH_3-N removal relationship

The data from the three trough experiments and two plot experiments were pooled to derive a generalised relationship between the rate of NH_3-N removal (kg NH_3-N / m^2 /d) and the leachate NH_3-N concentration. For each day of each experiment, the mass of NH_3-N removed was calculated and plotted against the leachate NH_3-N concentration at the start of that day (Figure 6). Figure 6 suggests that there is a relationship between the daily specific NH_3-N mass removal rate and the NH_3-N concentration. A linear regression line was fitted to the experimental data which indicates that there is a significant positive correlation ($p=0.01$) between the NH_3-N removal rate and NH_3-N concentration, although the regression line explains only 35% of the variation observed. This relationship could be used to simulate treatment plane performance and could form the basis of a methodology for system design. Such a methodology would need to take into account climatic factors that influence the recirculation reservoir volume, such as precipitation and evapotranspiration. Furthermore, the experimentally derived relationship should not be applied in practice without due consideration in the design process to the amount of variation in the rate of NH_3-N removal that has been shown to occur from batch to batch.

Fate of NH_3-N removed

A simple inorganic nitrogen balance was conducted by converting concentrations of NH_3-N, NO_2-N and NO_3-N at the beginning and end of the treatment plane experiments into values of mass, as presented in Table 5. In every experiment there was a significant reduction in the mass of inorganic nitrogen. NH_3 –N removal normally accounted for the majority of the inorganic nitrogen removed. NO_2-N removal contributed significantly to total inorganic nitrogen deficits in experiments C, D and E. NO_2-N is not normally a significant form of nitrogen in landfill leachate but its presence in this case was attributed to inputs of NO_2-N into the untreated leachate lagoon from a pilot leachate treatment plant operating at the Calvert landfill site. The mass of NO_3-N present in the treated leachate at the end of each experiment does not, however, account for the mass of NH_3-N and NO_2-N removed. This disparity between the removal of reduced forms of nitrogen and the consequent production of nitrate was also observed by Kruzic and Schroeder (1990) on domestic wastewater overland flow systems.

Kruzic and Schroeder (1990) argued that cation exchange of ammonium ions at soil particle surfaces would lead, in the short term at least, to the net removal of inorganic N from the water being treated. Regeneration of cation exchange sites would appear to be vital to the sustainable operation of a treatment plane, although in laboratory experiments it has been shown that cation exchange can facilitate considerable long-term NH_3-N removal in soils repeatedly challenged with leachate (Tyrrel, 1999). Gaseous nitrogen losses offer another explanation of the apparent deficit. Nitrate spike experiments carried out by Kruzic and Schroeder (1990) indicated that denitrification was probably occurring. Gaseous loss of ammonia to the atmosphere through the process of volatilisation may also occur. In studies of surface applied sewage sludge, Ryan and Keeney (1975) found that between 11% - 60% of applied ammoniacal nitrogen was lost by volatilisation, with the variation being dependent upon soil type. Soils with a relatively high cation exchange capacity, such as those used in the Silsoe experiments, would be expected to have relatively low volatilisation losses however. A further component of the apparent nitrogen deficit could be accounted for by assimilation of inorganic nitrogen into vegetation biomass. Irrigation of the Silsoe troughs and plots with leachate led to luxuriant growth of *Agrostis stolonifera*. Estimates made during the course of experiment E indicated that between 3-11% of the total nitrogen deficit for that experiment could be attributed to above ground biomass production. Laboratory experiments designed to verify the mechanisms responsible for NH_3-N removal and their relative significance have been conducted and will be reported elsewhere.

CONCLUSIONS

1. NH_3 -N removal from leachate applied to experimental treatment planes is a slow process in comparison with conventional wastewater treatment systems. An exponential relationship was found to best fit the experimental data (R^2 in the range 0.83-0.99).

2. The rate of NH_3-N removal was found to vary significantly between experimental runs. Although it is possible to identify possible causes of this variation it is not known if they could be brought under operational control in order to obtain more consistent performance. No relationship was observed between the hydraulic loading rate and the NH_3-N removal rate constants ($R^2 = 0.0039$).

3. The mass of NH_3 -N removed per square metre on any one day was linearly related to the NH_3 -N concentration at the start of that day of treatment as follows: NH_3 -N removal rate ($kg/m^2/d$) = 2×10^{-5} x NH_3 -N concentration (mg/l). This relationship could be used as a basis for a simple treatment plane simulation model. The development of such an application must take into account the considerable experimental variation observed and the need for the inclusion of climatic data so that changes in leachate volume may be catered for in the calculation.

4. The mass of NH_3-N and NO_2-N removed from the leachate applied cannot be accounted for by NO_3-N production in the final effluent. A combination of chemical and biological processes is likely to govern the fate of nitrogen in this system.

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Figure 1 Layout of the field-scale experimental treatment plane

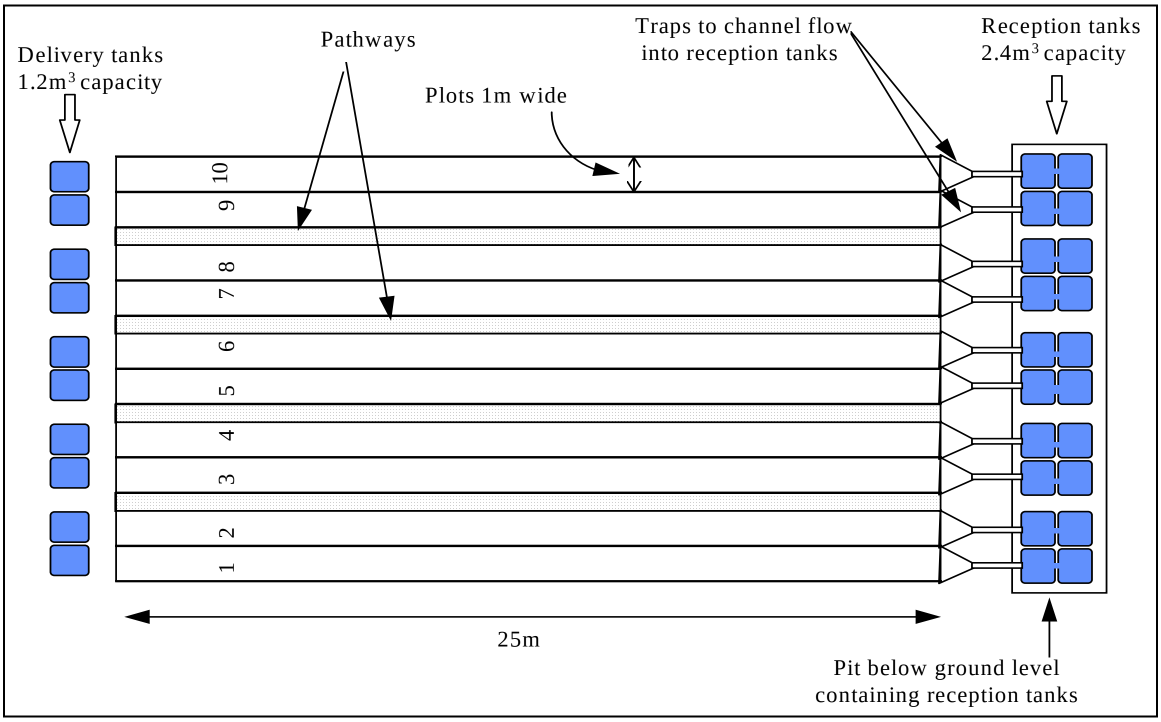

Figure 2 Diagram of a cross section of a pair of plots

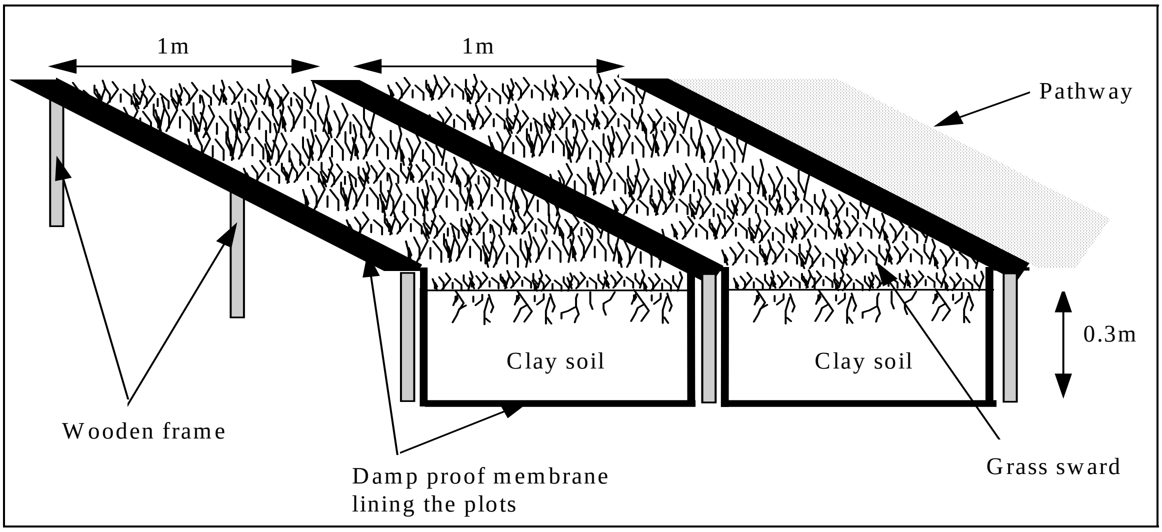

Table 1 Raw leachate quality from the Calvert landfill site over the duration of the study

Parameter	**Mean** (n=6)	**Maximum**	**Minimum**
pH	8.29	7.6	8.7
Chemical Oxygen Demand	1930 mg/l	700 mg/l	9304 mg/l
Chloride	3611 mg/l	2375 mg/l	6800 mg/l
Ammoniacal-N	514 mg/l	214 mg/l	812 mg/l
Nitrite-N	128 mg/l	1 mg/l	449 mg/l
Nitrate-N	27 mg/l	2 mg/l	95 mg/l

Figure 3 Schematic diagram of the trough experimental layout

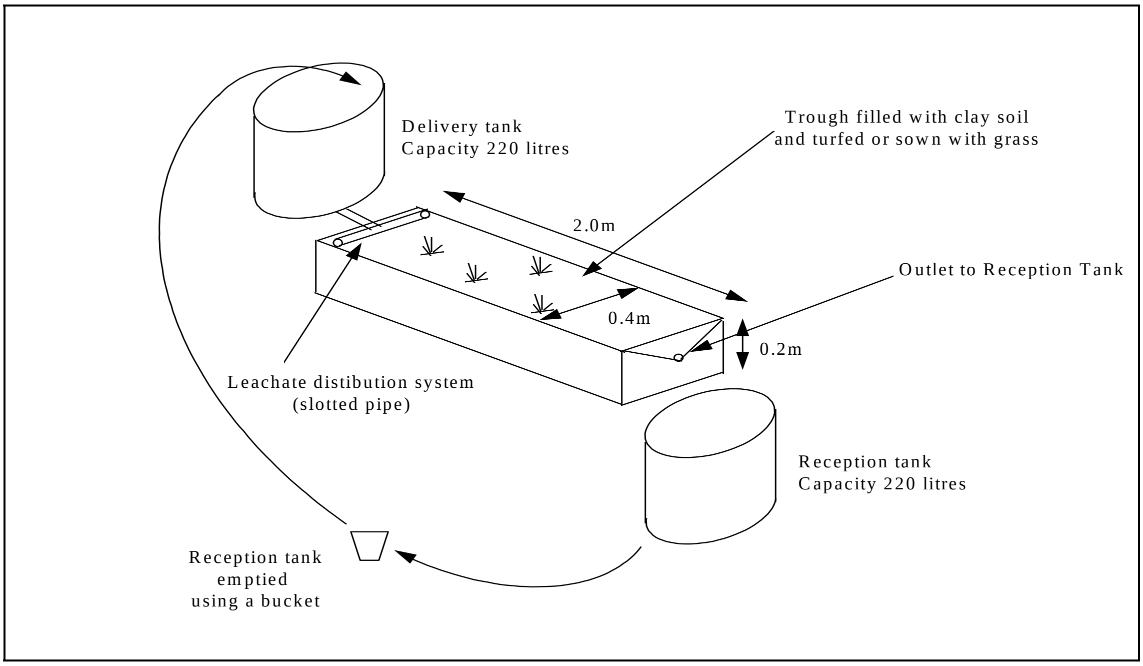

Table 2 Summary of the trough experimental programme

Experiment	Duration (days)	Trough number	Hydraulic loading rate ($l/m^2/d$)
A	32	Trough 1	87
		Trough 2	217
B	30	Trough 1	90
		Trough 2	44
		Trough 3	90
		Trough 4	44
C	18	Trough 1	78
		Trough 2	43
		Trough 3	17

NB: The hydraulic loading rate refers to the volume of leachate applied per square metre of treatment plane surface area during the 5 hours in the daily cycle that leachate was being applied.

Table 3 Summary of the plot experimental programme

Experiment	Duration (days)	Plot number	Plot length (m)	Hydraulic loading rate ($l/m^2/d$)
Start-up	13	Plots 1-10	25	Various
D	22	Plot 2	10	70
		Plot 6	20	24
		Plot 9	20	37
E	35	Plot 2	10	75
		Plot 4	10	45
		Plot 6	10	23
		Plot 9	10	45

NB: The hydraulic loading rate refers to the volume of leachate applied per square metre of treatment plane surface area during the 5 hours in the daily cycle that leachate was being applied.

Table 4 Best-fit exponential equations and coefficients of determination (R^2) for each of the experimental treatments.

Treatment identifier (experiment, plot/trough, number)	**Hydraulic loading rate ($l/m^2/d$)**	**Best-fit equation**	**R^2**
AT1	87	$495.48e^{-0.0535x}$	0.98
AT2	217	$454.47e^{-0.0479x}$	0.97
BT1	90	$439.09e^{-0.0575x}$	0.93
BT2	44	$346.82e^{-0.0273x}$	0.95
BT3	90	$149.71e^{-0.0787x}$	0.95
BT4	44	$142.59e^{-0.053x}$	0.97
CT1	78	$491.14e^{-0.0644x}$	0.99
CT2	43	$478.35e^{-0.0612x}$	0.99
CT3	17	$496.74e^{-0.0663x}$	0.98
DP2	70	$387.82e^{-0.071x}$	0.83
DP6	24	$486.63e^{-0.0985x}$	0.91
DP9	37	$364.39e^{-0.0507x}$	0.95
EP2	75	$139.68e^{-0.0412}$	0.85
EP4	45	$195.81e^{-0.0335}$	0.92
EP9	45	$237.48e^{-0.0463x}$	0.90
EP6	23	$249.56e^{-0.0382x}$	0.90

Figure 4 A typical example of NH_3-N removal from a batch of leachate (experiment A, small–scale trough 1)

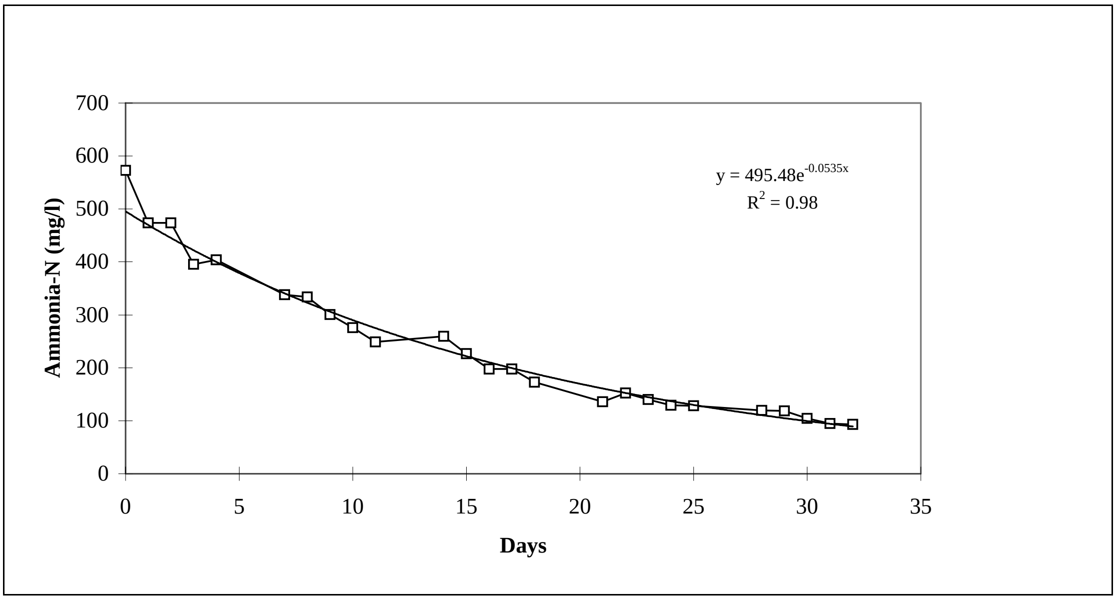

Figure 5 The relationship between hydraulic loading rate and the ammoniacal nitrogen removal rate constant (k)

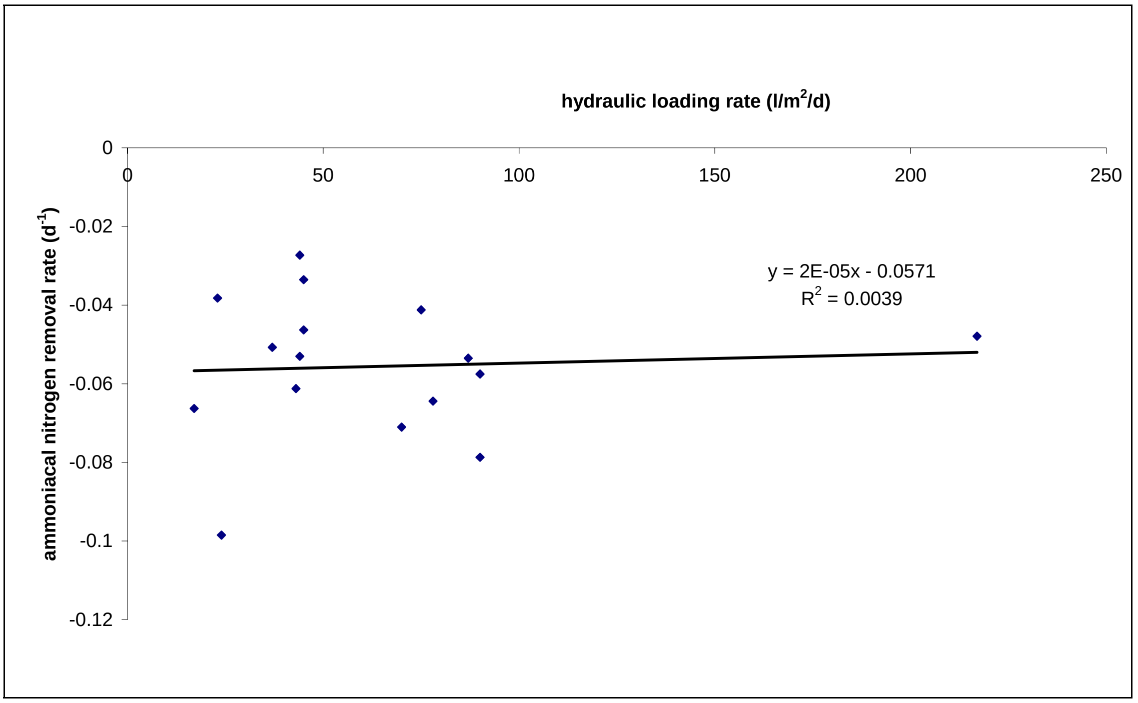

Figure 6 Relationship between NH_3-N removal rate and concentration for pooled experimental data.

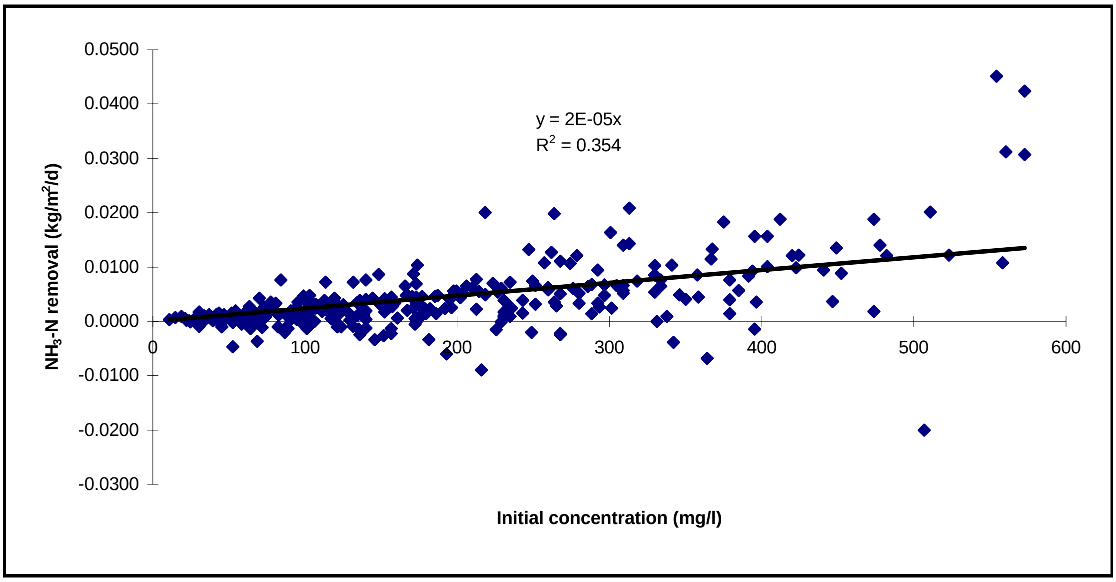

Table 5 Mass of nitrogen in leachate at the beginning and end of experiments A-E

Treatment identifier (experiment, plot/trough, number)	**Total mass of inorganic N (kg) in leachate**		**% of inorganic N remaining at end**	**% of change in inorganic N accounted for by changes in:**		
	Beginning	**End**		**NH_3-N**	**NO_2-N**	**NO_3-N**
AT1	0.1422	0.0263	18	88	7	5
AT2	0.1422	0.0291	20	88	8	4
BT1	0.0293	0.0073	25	104	5	-9
BT2	0.0334	0.0107	32	103	6	-9
BT3	0.0925	0.0105	11	94	5	2
BT4	0.0852	0.0268	31	88	6	6
CT1	0.1342	0.0341	25	84	17	-2
CT2	0.1351	0.0392	29	87	17	-4
CT3	0.1401	0.0606	43	89	15	-4
DP2	1.587	0.0111	1	83	17	0
DP6	1.631	0.0512	3	83	19	-2
DP9	1.5379	0.0441	3	81	20	-1
EP2	1.5668	0.0409	3	33	65	2
EP4	1.7058	0.3877	23	41	55	4
EP6	1.7699	0.5063	29	53	45	2
EP9	1.5096	0.2408	16	42	55	3