Pergamon

PII: S0043-1354(97)00392-8

Wat. Res. Vol. 32, No. 5, pp. 1383–1392, 1998

Printed in Great Britain
0043-1354/98 $19.00 + 0.00

IN SITU NITROGEN MANAGEMENT IN CONTROLLED BIOREACTOR LANDFILLS

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(First received January 1997; accepted in revised form September 1997)

Abstract—The characteristics of leachate from landfills vary according to site-specific conditions. Leachates from "old" landfills are often rich in ammonia nitrogen due to the hydrolysis and fermentation of the nitrogenous fractions of biodegradable substrates, with decreases in concentration mainly attributable to leachate washout. At landfills where leachate containment, collection and recirculation is practiced to accelerate decomposition of readily biodegradable organic constituents, leachate ammonia nitrogen concentrations may accumulate to higher levels than during conventional single pass leaching, thereby creating an ultimate leachate discharge challenge. Landfill leachate treatment options include complex and often costly sequences of external physical–chemical and biological processes for removal of high-strength organics and inorganics, including nitrogen. Therefore, this paper focuses on investigations with bioreactor landfill simulations to demonstrate the potential for *in situ* nitrogen removal in dedicated nitrification/denitrification zones. Using leachate recirculation, associated system modifications provided separate aerobic and anoxic zones for ammonia nitrogen transformations to nitrate and nitrogen gas, respectively. Results from the three simulated optional stages of methanogenesis, nitrification and denitrification indicated that nitrogen conversion and removal was dependent on the operational stage. Both separate and combined reactor operation with internal leachate recycle provided 95% nitrogen conversion. In contrast, combined reactor operation with single pass leaching provided a conversion efficiency per cycle ranging between 30–52% for nitrification and 16–25% for denitrification, thereby indicating the efficacy of using the landfill itself for attenuation of leachate ammonia nitrogen concentrations to levels acceptable for ultimate discharge. © 1998 Elsevier Science Ltd. All rights reserved

Key words—landfill bioreactor, leachate recirculation, landfill stabilization, nitrogen removal, nitrification, denitrification

INTRODUCTION

Leachates from the final maturation phase of landfill stabilization are often rich in ammonia nitrogen due to the hydrolysis and fermentation of the nitrogenous fractions of biodegradable substrates (Dedhar and Mavinic, 1986; Carley and Mavinic, 1991). Moreover, at landfills where leachate containment, collection, and recirculation are practiced to accelerate decomposition of readily degradable organic constituents, leachate ammonia nitrogen concentrations may accumulate to higher levels than during conventional single pass leaching, thereby requiring treatment prior to ultimate discharge (Pohland, 1995).

External treatment options for landfill leachate may require physical–chemical and biological processes for removal of high-strength organic and inorganic materials, including nitrogen (Keenan *et al.*, 1984). Biological treatment of leachate with high ammonia concentrations has been investigated by a number of researchers (Knox, 1985; Carley and Mavinic, 1991; Hosomi, 1991; Opatken and Bond, 1991; Monoharan *et al.*, 1992). If provided by a separate treatment system, additional costs and operational challenges may result. Because leachate recirculation through a landfill accelerates refuse stabilization and enhances gas production, there is current interest in its application in practice. Therefore, this research was conducted with laboratory-scale simulated landfill units to demonstrate the feasibility of extending this stabilization to *in situ* nitrification and denitrification at controlled landfills operated with leachate recycle.

CONCEPTUAL CONFIGURATION

The landfill environment is a complex heterogeneous system in which different types of microorganisms coexist. These microbial populations are capable of a variety of reactions, depending upon the prevailing environmental conditions and the organisms-substrate specificity (Ragle *et al.*, 1995).

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Because biological nitrification–denitrification is a commonly used wastewater treatment technology for nitrogen removal, a three-component simulated landfill system was developed to include anoxic, anaerobic and aerobic zones as illustrated in Fig. 1. This configuration facilitated the integration of anaerobic degradation, with methane and carbon dioxide generation and two separate aerobic and anoxic zones for nitrogen conversion and removal (Onay, 1995). These modifications to the usual design and operation of a bioreactor-type landfill were intentionally incorporated for *in situ* attenuation of high residual leachate ammonia nitrogen concentrations characteristic of the final maturation phase of landfill stabilization.

Leachate recycle was used to transport stabilization products from one landfill layer to the next and thereby enhance conversion on a continuum. The sources of carbon and nitrate necessary for denitrification could be supplied by utilizing leachate recycle to carry the residual C and N from the anaerobic zone into the aerobic zone, and subsequently to the anoxic zone at the top of the system. Accordingly, leachate nitrogen could be removed concomitantly with other attenuation, e.g., sulfur compounds (Onay and Pohland, 1995), without the need for external treatment prior to ultimate disposal.

MATERIALS AND METHODS

Simulated landfill bioreactor construction

To demonstrate the utility of the modified landfill system and to meet project objectives, a three-reactor system designed to simulate the landfill environment and to investigate the potential for *in situ* attenuation of nitrogen compounds was constructed with lengths of Schedule 40 PVC. The larger methanogenic reactor had a diameter of 0.4 m and length of 1 m, while the diameter and length of the smaller nitrification and denitrification reactors were 33.02 and 60.96 cm, respectively. The configuration and oper-

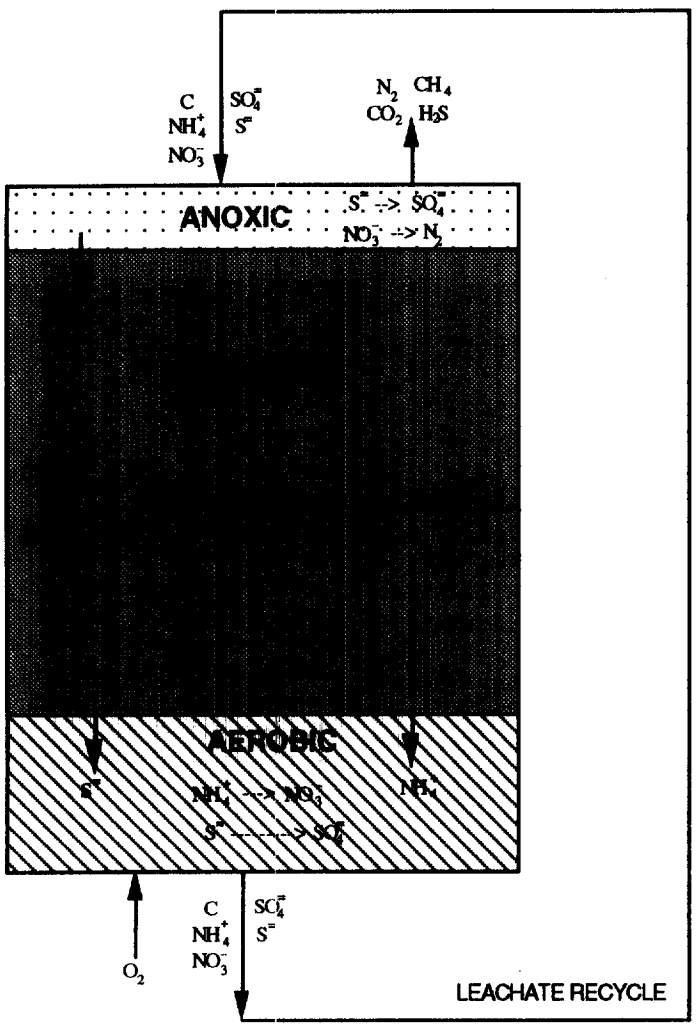


Fig. 1. Conceptual landfill configuration for *in situ* nitrification and denitrification.

ational features of the reactors are illustrated in Fig. 2. During the nitrification study, the air inlet at the bottom of the reactor was connected to an air pump operating at 93 liters oxygen per day flow rate to maintain aerobic conditions. However, when the methanogenetic and denitrification reactors were used, the air inlet was closed in order to develop anaerobic and anoxic conditions, respectively. PVC flanges were used both at the top and bottom of the reactors to provide support for the top and bottom lids.

Simulated landfill bioreactor loading

Compost was utilized as the waste matrix in the simulated landfill bioreactors because of its similarity to the waste mass present in landfills after accelerated stabilization and the final maturation phase of landfill stabilization had been reached. Compost has a relatively lower available carbon content than municipal refuse, whereas its nitrogen content is higher. In addition, the presence of trace minerals, i.e., calcium, magnesium, manganese, iron and potassium, helped to assure environmental conditions conducive to the rapid development of the microbial populations in the reactors (Table 1). Pine bark chips were also used as a bulking agent in all three reactors to promote more uniform gas exchange and distribution, particularly of air from the bottom to the top of the waste matrix in the nitrification reactor. The same mixture of

Table 1. Characteristics of compost used in landfill bioreactor simulations

Parameter	Results[a]
Total Kjeldal nitrogen (TKN)	1.31
Ammonia nitrogen (NH_3N)	0.16
Nitrate nitrogen (NO_3N)	<0.01
Phosphorus (P_2O_5)	1.15
Calcium (Ca)	2.1
Magnesium (Mg)	0.37
Manganese (Mn)	0.065
Iron (Fe)	1.4
Potassium (K_2O)	0.28
Carbon (C)	33.2
Alkalinity ($CaCO_3$)	5.58
Total solids	72.5
Volatile solids	60
Wet density (kg/m^3)	502

[a]Results are expressed in percent by weight of waste matrix added, unless an alternative unit is given.

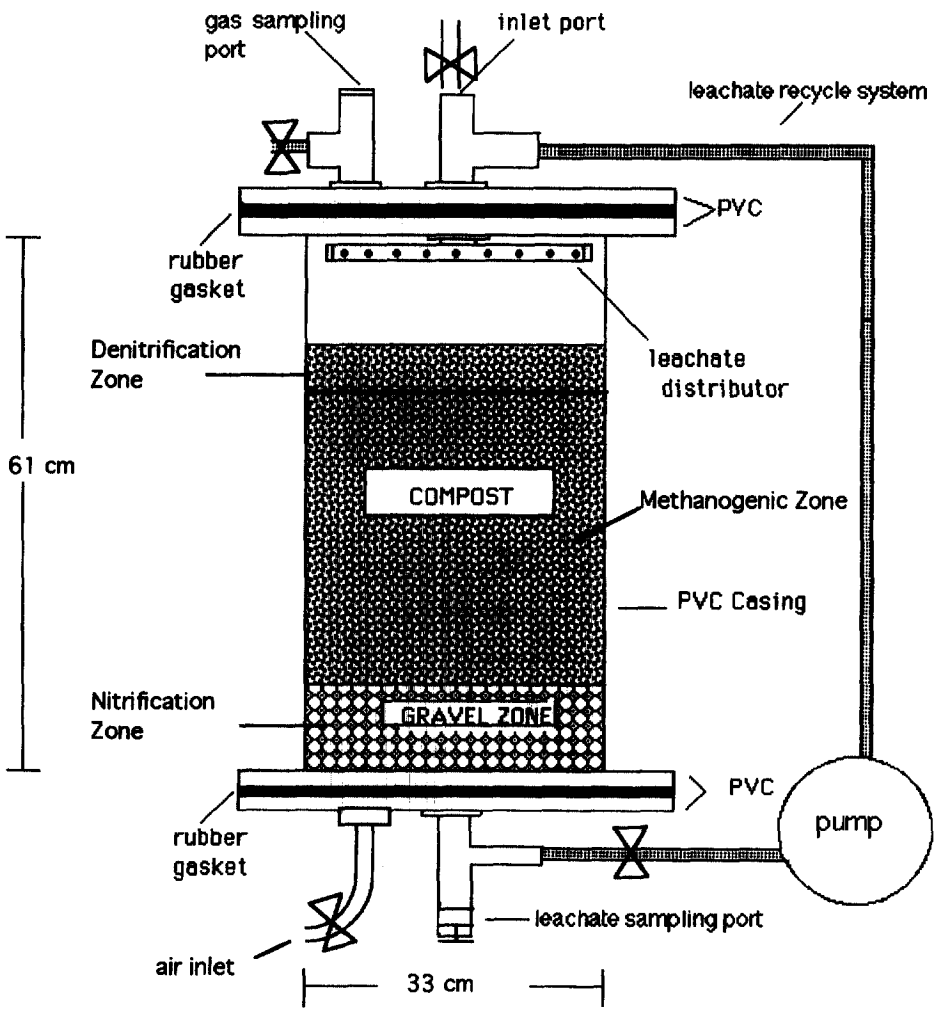


Fig. 2. Configuration and operational features of the landfill bioreactor for *in situ* nitrification and denitrification.

compost and bulking agent was used in all three reactors to provide consistency of the waste matrix. Upon closure, the PVC flange and the top lid of each reactor were sealed and, with the exception of the nitrification reactor, the reactors were purged with nitrogen gas, connected to a gas collection unit, and placed in an incubator maintained at a constant temperature of 37°C.

Analytical methods

In order to help establish the extent or condition of nitrification, denitrification and methanogenesis, leachate samples were collected from each reactor and analyzed for chemical oxygen demand (COD), volatile organic acids (VOAs) including acetic, propionic, *iso*-butyric, butyric, *iso*-valeric, valeric, and caproic acids, pH, oxidation–reduction potential (ORP), ammonia nitrogen, and anions including chloride, bromide, nitrate, phosphate, sulfate. A gas chromatograph was used for VOA analysis, and anions were analyzed with an ion chromatograph. Gas production and composition by gas chromatography were also monitored on a daily basis. All other analyses were conducted according to *Standard Methods* (1985).

Simulated landfill bioreactor operations

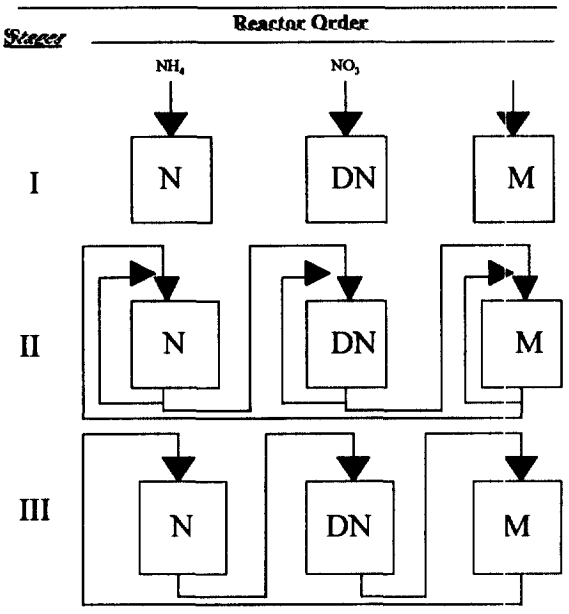
Because the reactor experiments involved the final maturation phase of landfill stabilization and the conversion and disposition of nitrogenous compounds under various potential modes of operation, the experimental effort was divided into three operational stages; separate reactor operation, combined reactor operation with internal leachate recycle, and combined reactor operation with single pass leaching (Table 2). The main purpose of conducting the separate reactor stages was to ensure the initiation and development of respective microbial activity, i.e., denitrification, nitrification, and residual methanogenesis. Therefore, each reactor environment was modified, operated, and monitored to allow the onset of these conditions, and batch chemical additions of ammonia and nitrate were introduced to the nitrification and denitrification reactors, respectively, in order to facilitate the desired microbially-mediated nitrogen conversion.

After establishing active microbial populations in each, the reactors were connected in sequence to simulate leachate recirculation patterns that would occur in a landfill with dedicated treatment zones (Fig. 1). Therefore, this stage involved combined operation with leachate recycle around each reactor. The initial operation of this second stage was commenced with transfer of the leachate from the denitrification reactor to the methanogenic reactor. In addition, the liquid content of the nitrification reactor was transferred to the denitrification reactor for the removal of available nitrate in the leachate prior to its subsequent transfer also to the methanogenic reactor.

After accumulating the liquid content of all reactors in the methanogenic reactor, the leachate transfer cycles between reactors were begun. A total of nine cycles were completed between the reactors in order to demonstrate attenuation and conversion potentials for the leachate nitrogen. Leachate with high leachate ammonia nitrogen concentration from the methanogenic reactor was first cycled through the nitrification reactor where the conversion of ammonia nitrogen to nitrate was achieved. The resulting effluent was then cycled through the denitrification reactor for conversion of nitrate to nitrogen gas. Finally, the liquid content of the denitrification reactor was transferred to the methanogenic reactor which functioned to convert residual nitrate to ammonia and as a storage location for the leachate until the next cycle was initiated. In addition to the external transfer cycles, each reactor received internal recycle of leachate once a week during this stage to accelerate and complete the desired conversion of nitrogenous substrate.

During the third stage of operation, the transfer of the liquid content from one reactor to another was performed without employing internal leachate recycle around each reactor (Table 2). This stage was undertaken in order to more closely simulate conventional landfill leachate flow patterns. A total of 14 cycles were conducted for each reactor within an operational period of 20 days. After its introduction to the reactors, leachate was allowed to travel through the waste matrix to the bottom of the reactors for collection, a travel time of 30 to 45 min. As a result, the conversion of the nitrogenous compounds during each pass was limited by the retention time in the reactors and the associated contact opportunity with the indigenous microorganisms. Conversion and removal of the nitrogenous substrate were determined after each cycle, and sequential cycles between reactors were employed to more fully investigate the possible extend of the attenuation and removal under single pass leaching conditions and without internal leachate recycle.

Table 2. Operational modes of reactor system



N: nitrification; DN: denitrification; M: methanogenic.

RESULTS AND DISCUSSION

Table 3 provides an example of changes in some of the other process variables for the first operational stage. High chemical oxygen demand (COD) removal was observed in both the denitrification and nitrification reactors. Gas production rate was temporarily high at the beginning of the study due to anaerobic stabilization of residual carbon from the compost, with methane being the major gas component. However, after the onset of denitrification, methane was displaced by nitrogen gas. The following sections detail associated nitrogen transformations throughout the three experimental stages.

Table 3. Example changes in process variables; first operational stage

Parameter	Denitrification reactor		Nitrification reactor	
	initial	final	initial	final
COD (mg/l)	13000	3000	14000	1000
pH	7	7.6	7.1	6.3
ORP (mv)	−50	−250	—	—
TVOA (mg/l) as acetic acid	800	10	—	—
Total gas production (l)	0	25	—	—
Methane production (l)	0	5	—	—
Carbon dioxide production (l)		10	—	—

Nitrogen transformations during the first stage of operations

During the first stage operation of the denitrification reactor, 16 batches of potassium nitrate (KNO_3), in concentration equivalent to the ammonia produced during the anaerobic decomposition of compost, were prepared and introduced intermittently to simulate leachate recycle sequences. As indicated in Fig. 3, during the first 120 days when methanogenesis occurred, nitrate was temporarily converted to ammonia nitrogen, and nitrogen gas decreased due to gas displacement by methane and carbon dioxide. However, after initiation of denitrification, nitrate concentrations were reduced from 750 to 1 mg/l within 3 to 5 days after each batch addition of KNO_3. Correspondingly, an increase in nitrogen gas (up to 65%) was observed.

For the nitrification reactor, seven batches of ammonium chloride were introduced during the first stage of operations, yielding an average ammonia nitrogen concentration of 150 mg/l. The complete conversion of ammonia nitrogen to nitrate within less than a week was observed after each batch addition (Fig. 4). The relatively high leachate ammonia nitrogen concentration initially measured was due to the washout of ammonia from the compost. After oxidation of this initial ammonia and the onset of nitrification within 20 days after the start of operations, the batches of ammonium chloride introduced to simulate leachate recycle sequences were completely converted to nitrate. In terms of overall mass, cumulative amounts of 6.0 g of nitrate nitrogen and 1.5 g of ammonia nitrogen were loaded to the denitrification and nitrification reac-

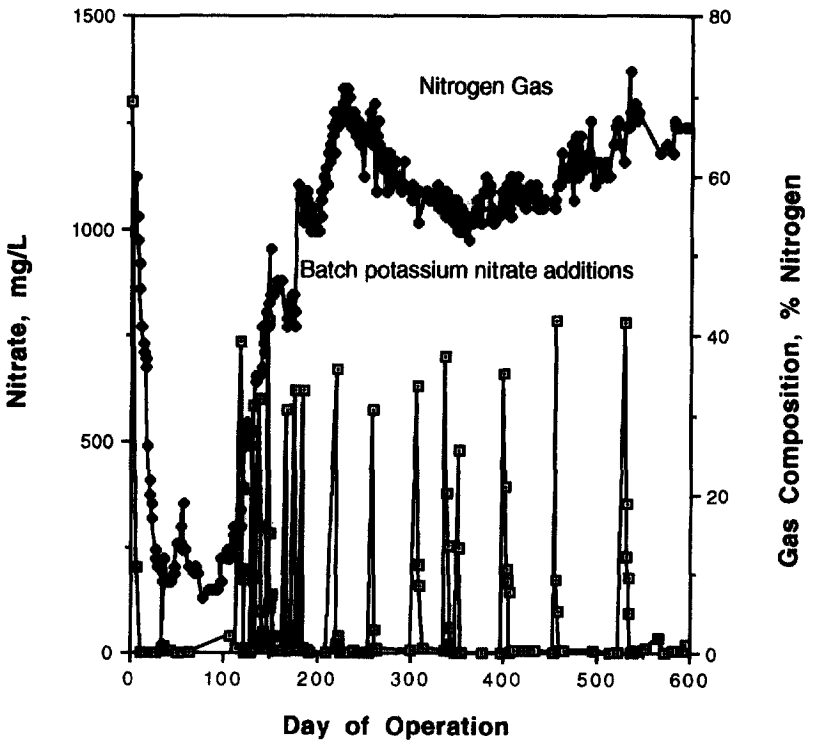


Fig. 3. Leachate and gas-phase nitrogen concentrations from the denitrification reactor.

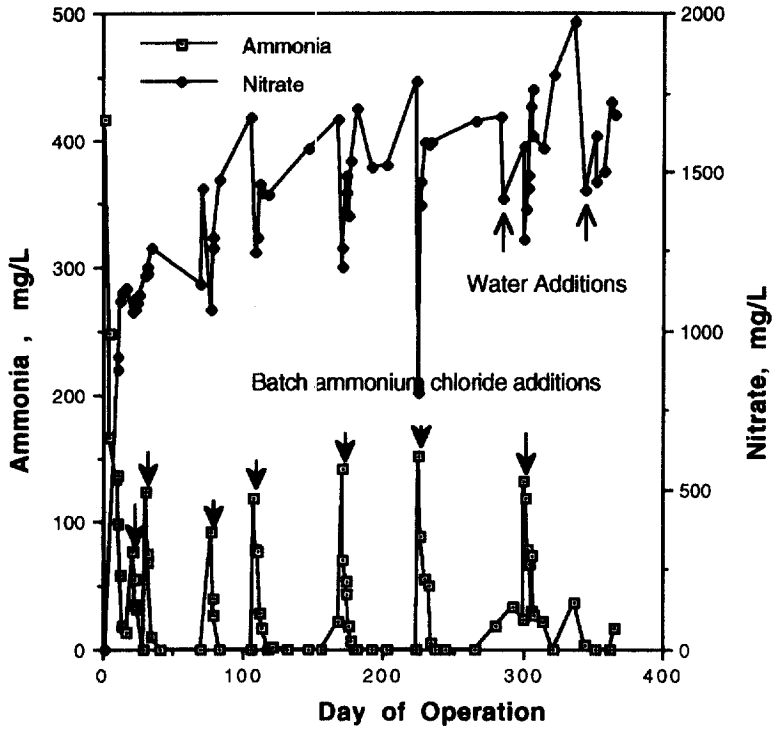


Fig. 4. Leachate nitrogen concentrations from the nitrification reactor.

tors, respectively, and nitrogen recoveries from both reactors were higher than 95%.

Nitrogen transformations during the second stage of operations

After initiation of active nitrification and denitrification during the first stage, the reactors were connected to simulate the leachate flow sequences indicated in Fig. 1. In this stage, however, internal leachate recycle around each reactor was provided to facilitate the more rapid and complete conversion of nitrogenous substrate. Figure 5 illustrates the conversion of nitrate to nitrogen gas by denitrification for the cycles during the second stage of operations. A nitrate peak resulted for each cycle after introduction of the high nitrate leachate from the nitrification reactor. Leachate nitrate concentrations, ranging from 920 to 1400 mg/l, were rapidly converted to nitrogen gas within about 10 days of operation. The actual nitrogen gas concentration in the gas phase was increased from 66.5% to approximately 74% as a result of the nitrate reduction. Moreover, the recycling of leachates between bioreactors increased the nitrogen load to the denitrification reactor from one cycle to another. Despite this increase, denitrification proceeded rapidly, resulting in sharp decreases in leachate nitrate concentrations. The cumulative nitrogen mass of 1.05 g applied to the denitrification reactor resulted in a final nitrogen mass recovery of 94%. Nitrogen removal via denitrification ranged from 91% to 93% throughout the second stage of operation.

Nitrogen conversions in the nitrification reactor during the second stage of operations are illustrated in Fig. 6. Initial leachate ammonia concentrations of 450 and 512 mg/l, respectively, resulted for Cycle 1 and 2 due to the introduction of leachate from the methanogenic reactor. This ammonia nitrogen concentration in the leachate was equivalent to a nitrogen mass of 0.509 g, which was five times higher than in the ammonium chloride batches that were introduced during the first stage of operation of the nitrification reactor. However, conversion under this high loading was very reliable, with almost 99% of the ammonia nitrogen being transformed to nitrate by nitrification. The ammonia nitrogen was almost completely converted within ten days, with a concomitant increase in leachate nitrate concentration from 1.400 mg/l to approximately 2.800 mg/l.

Nitrogen transformations during the third stage of operations

Figures 7–9 indicate the change in the mass of the various nitrogen species for the nitrification, denitrification, and the methanogenic reactors throughout the third stage of operation. As indicated in the figures, two bars represent each cycle

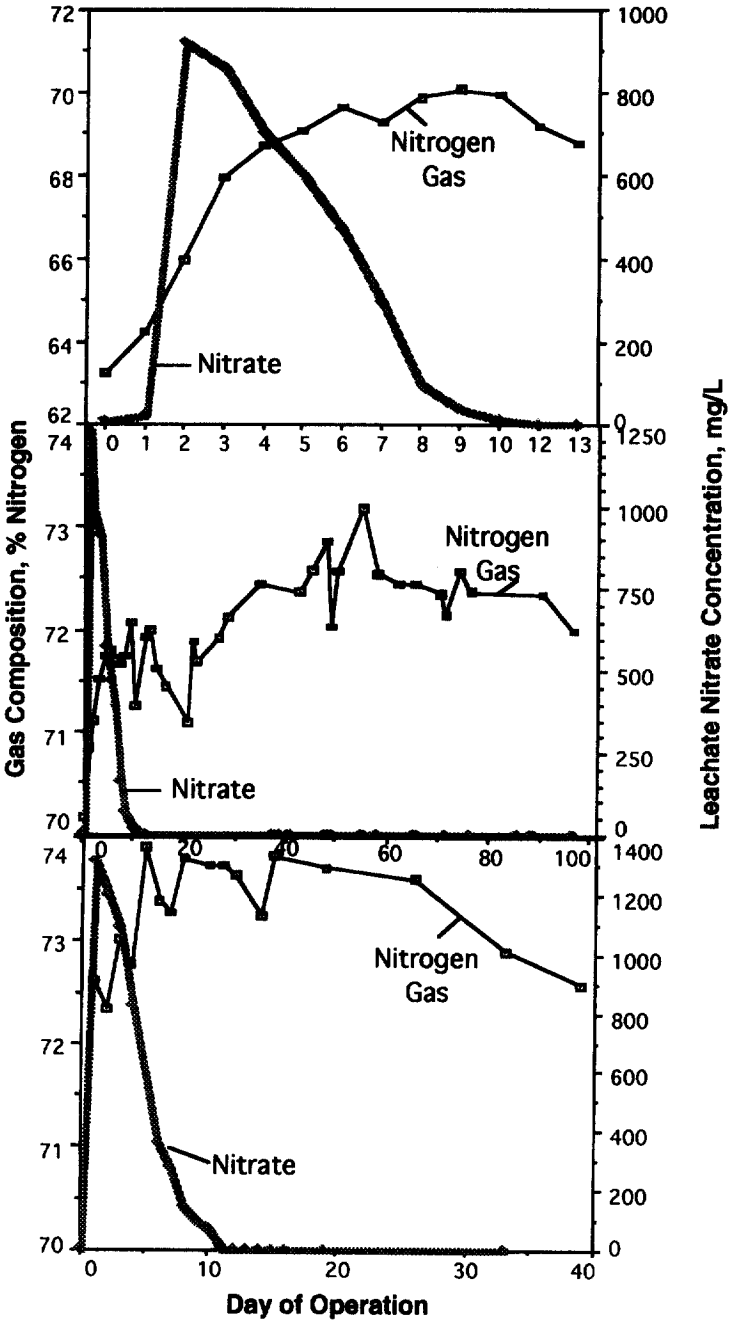


Fig. 5. Leachate and gas-phase nitrogen concentration from the denitrification reactor during the second stage of operations.

for each reactor. The first and the second bar for each cycle indicate the mass of nitrogen entering and exiting the reactor, respectively. Due to the more saturated and uniform environment created by the moisture additions for each reactor during the third stage of operation, the mass of nitrogen entering was equivalent to the mass exiting the reactor, as is indicated in the figures with bars of the same height for corresponding cycles.

Figure 7 illustrates the change in nitrogen mass for the nitrification reactor throughout 14 cycles. The ammonia nitrogen generated in the methanogenic reactor was loaded to the nitrification reactor, where conversion to nitrate nitrogen was about 30% per cycle. Similar to the trend observed for the nitrification reactor, as the sequential cycles between the bioreactors progressed, the nitrogen loading to the denitrification reactor decreased along with the

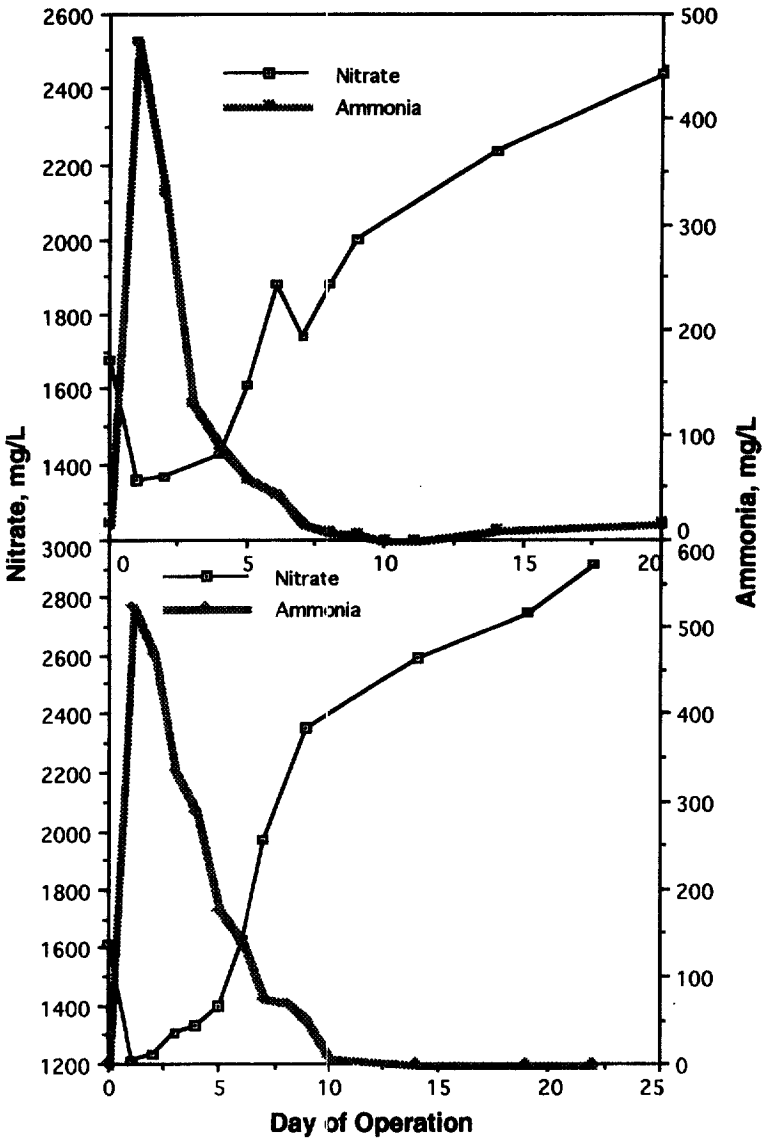


Fig. 6. Leachate nitrogen concentrations from the nitrification reactor during the second stage of operations.

increase in the nitrogen gas conversion efficiency (Fig. 8). The total nitrogen loading was decreased from 0.8454 to 0.1173 g at the end of Cycle 14, and the denitrification efficiency at the end of each cycle had increased from 6.16% to 27% due to the decrease in the nitrogen loading applied to the denitrification reactor. Therefore, by controlling the nitrogen loading and the associated leachate recirculation to the system, the denitrification efficiency could be improved. The results from the denitrification reactor indicated that an average conversion efficiency of 15.75% was achieved after each cycle. Meanwhile, the unconverted portions of nitrate nitrogen was partially reduced to ammonia nitrogen in the methanogenic reactor under the existing reducing environment (Fig. 9), and was then used as an ammonia nitrogen source for nitrification in the succeeding cycle.

SUMMARY AND CONCLUSIONS

A landfill bioreactor system was designed and operated with sequential *in situ* removal of the nitrogenous substrates in dedicated nitrification and denitrification zones. The extent of stabilization obtained, as reflected by the gas generated and selected leachate indicator parameters, emphasized the beneficial effect of leachate recirculation on the performance of the simulated landfill system. Utilization of leachate recirculation enhanced and

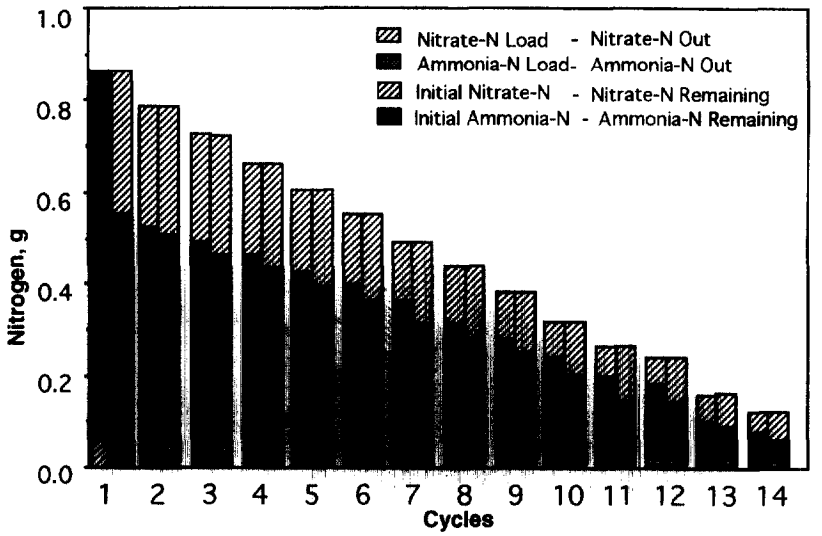


Fig. 7. Mass of cumulative leachate nitrogen from the nitrification reactor during the single pass operational stage.

accelerated conversion and stabilization in all of the reactors by increasing the uniformity of moisture, substrate and nutrient distribution. Consequently, leachate recirculation created an environment that promoted the rapid development of the desired microbial populations of denitrifiers, nitrifiers, and methanogens.

The efficiency of nitrogen conversion was dependent on the operational stages, and both separate and combined reactor operation with internal leachate recycle around each reactor provided 95% nitrogen conversion. In contrast, combined reactor operation without internal recycle provided a conversion efficiency per cycle ranging between 30–52% for nitrification and 16–25% for denitrification, with a suggestion of increasing trends as acclimation became better established. Because the feasibility of *in situ* nitrification and denitrification in a bioreactor landfill was demonstrated, design modifications involving an aerobic zone associated with the leachate underdrain system, and an anoxic zone associated with a surfacial leachate distribution system below the final cap, or *vice versa* are recommended.

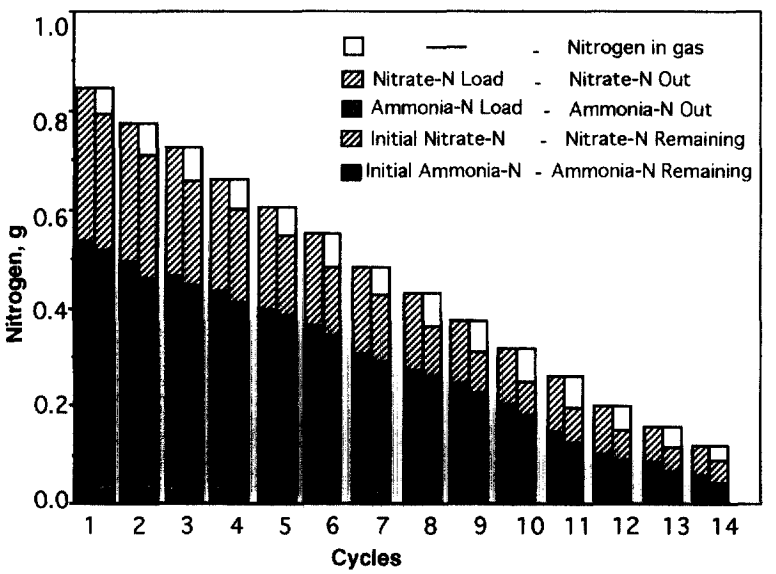


Fig. 8. Mass of cumulative leachate nitrogen from the denitrification reactor during the single pass operational stage.

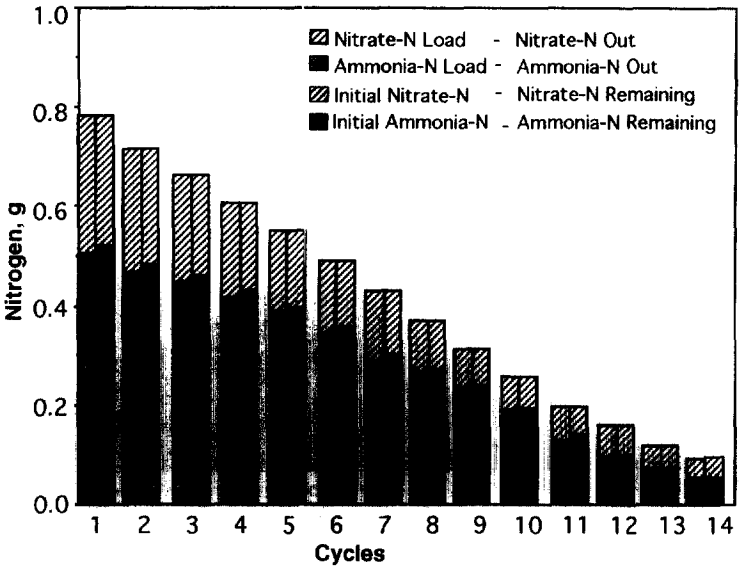


Fig. 9. Mass of cumulative leachate nitrogen from the methanogenic reactor during the single pass operational stage.

Acknowledgements—The authors acknowledge the financial support provided by the Delaware Solid Waste Authority and the Turkish Ministry of Education throughout the conduct of this research.

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