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Nitrogen removal from recycled landfill leachate by ex situ nitrification and in situ denitrification

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Abstract

A three-compartment system, comprising a landfill column with fresh municipal solid waste, a column with a well-decomposed refuse layer as methane producer, and a sequential batch reactor as ex situ nitrifying reactor, was employed to remove nitrogen from municipal solid waste leachate. Since food waste comprised a major portion of refuse collected in Shanghai, an intense hydrolysis reaction occurred and caused the rapid accumulation of ammonia nitrogen (NH_3–N) and total organic carbon in the leachate. This paper discusses the role of the three mentioned units and the design and operation of the proposed system. With most NH_3–N being converted to nitrite nitrogen (NO_2^-–N) or nitrate nitrogen (NO_3^-–N) by the nitrifying reactor, and with the well-decomposed refuse layer transforming most dissolved organic compounds to CO_2, carbonates and methane, it was found that the fresh refuse column could efficiently denitrify the hydrolyzed nitrogen to N_2 gas. The role of the three mentioned units and comments on the design and operation of the proposed system are also discussed.

1. Introduction

In addition to providing accelerated stabilization of landfilled municipal solid waste (MSW), a landfill bioreactor can also enhance methane production, thus presenting a viable alternative for urban waste management (Mehta et al., 2002; Reinhart et al., 2002; Barlaz and Reinhart, 2003). However, the recycling of leachate inevitably causes ammonia nitrogen (NH_3–N) accumulation, which is almost inert under anaerobic conditions (Pohland, 1995). This high-strength wastewater requires intensive treatment to enable safe disposal. Physical–chemical or biological processes are generally adopted for ex situ treatment of leachate (Keenan et al., 1984).

Onay and Pohland (1998) presented a laboratory-scale test in simulated landfill units to demonstrate the feasibility of removing nitrogen by use of leachate recycling. These authors devised a novel process with minor aeration at the landfill base for converting NH_3–N into nitrate nitrogen (NO_3^-–N). This nitrate-rich stream was recycled and denitrified at the anaerobic zone. The proposed in situ nitrification–denitrification process appeared more promising than the conventionally adopted ex situ processes owing to its lower operational cost and lower space requirements. However, since oxygen and nitrogen gas were conveyed into the landfill layer, the methane concentration in the biogas was reduced. Jokela et al. (2002) refined the work of Onay and Pohland (1998) by utilizing an ex situ biofilter for nitrification. Burton and Watson-Craik (1999) operated a landfill test cell designed to denitrify externally nitrified leachate. It was shown that the NO_3^-–N returned to the landfill cell was consumed under the anoxic/anaerobic landfill conditions. Price et al. (2003) fed 400 mg NO_3^-–N/L into a series of reactors that were filled with fresh and well-decomposed waste to investigate the process of denitrification in the reactors, and its influence on methane production. The study demonstrated that NO_3^-–N was effectively converted to nitrogen when

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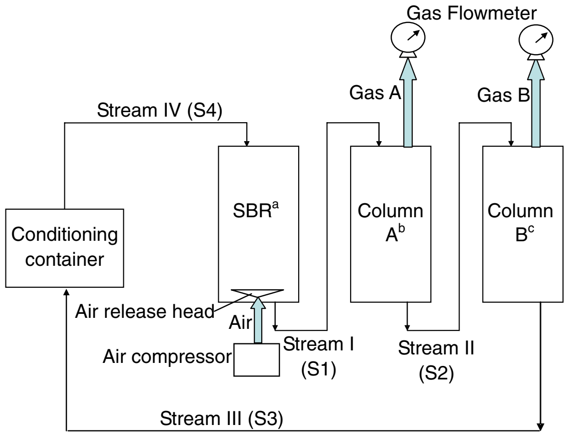
supported with adequate organic carbon as an electron donor, while methane production was inhibited in the process. Since the composition of landfill leachate is more complicated than that of a NO_3^-–N solution, and includes other components besides NO_3^-–N, the denitrification capacity of landfilled waste to the recycled-leachate should be further examined. Berge and Reinhart (2003) conducted a study evaluating the kinetics of in situ nitrification removal processes in landfills with air addition, and the results suggested that denitrification could occur quickly in the landfill layer.

This investigation refines the combined process devised by Pohland and co-workers for nitrifying the NH_3–N of leachate in an external reactor and denitrifying the yielded nitrite nitrogen (NO_2^-–N) or NO_3^-–N to nitrogen gas in a well-decomposed waste layer. Unlike previous studies, the fresh refuse column tested in this study was supplemented with a well-decomposed refuse layer as the in situ methane producer. The refuse used in this study was collected in Shanghai and reflected typical waste from the city. This study is the first to report the use of a combined ex situ nitrification and in situ denitrification process for managing nitrogen in leachate from fresh refuse collected in China.

2. Experimental

The combined nitrification–denitrification–methanogenesis system involved a nitrifying reactor, a simulated landfill column loaded with fresh synthetic waste, and another simulated landfill column loaded with well-decomposed landfilled waste (Fig. 1).

The leachate was nitrified in the nitrifying reactor before it was fed into Column A (Stream I, S1). The leachate produced from Column A was then fed into Column B via Stream II, S2. Finally, the leachate from Column B (Stream III, S3) was recycled back via a conditioning container to the nitrification reactor to create a closed loop. The gas streams from Columns A and B were named as "Gas A" and "Gas B", respectively. All experiments were performed in a thermostatic chamber and the temperature was kept at 35 ± 2 °C. (The temperature in a Chinese landfill ranges from 22 to 52 °C, and is 27–45 °C over the first 2 yr after landfilling. The temperature of 35 °C is about the mean temperature over the temperature range noted in field.)



[a] sequential batch reactor (SBR) served as the nitrifying reactor

[b] loaded with the synthetic refuse and served as a hydrolysis and denitrification reactor

[c] loaded with well-decomposed waste and served as an organic carbon consumption reactor

Fig. 1. Schematic of the combined nitrogen removal process.

Columns A and B were made of plexiglass (diameter 19 cm and height 90 cm). The synthetic refuse loaded in the simulated landfill columns was prepared as described in a survey on MSW composition in Shanghai (He, 2002; He et al., 2005). Table 1 lists the composition and characteristics of the synthetic refuse. Food waste were from the refectory of Tongji University, and plastics, paper, textiles, glass and metal were hand-picked from the MSW in a waste transfer station in the urban area of Shanghai. All of the waste fractions were then coarsely shredded (<3 cm range to avoid preferential flow) and commingled by hand. The refuse composition was expected to emulate the average composition of typical landfilled municipal solid waste in China, mainly composed of biodegradable wastes. The synthetic refuse was packed into Column A at a density of 800 kg/m^3.

The well-decomposed waste, excavated from a cell in the Laogang MSW Landfill in Shanghai, whose final cover system had been installed for about 5 yr, was loaded in Column B. The ratio of cellulose/lignin of the refuse was less than 0.5, and the composition was mainly plastics and slag with the color of black. The initial packing density of the well-decomposed waste was 1100 kg/m^3.

The effluent leachate of the nitrifying reactor was then pumped to Column A at a rate of 18.7 mm/d, i.e., 420 ml, for 150 days. Thereafter, the leachate-recycling rate was adjusted to 11.2 mm/d, i.e., 250 ml, to reduce the leachate production rate during the later phase of stabilization.

The nitrifying reactor was made of plexiglass (diameter of 14 cm) and had an effective volume of 9 L. Compressed air was generated by an air compressor and was introduced to the air release head located at the bottom of the nitrifying reactor. The reactor was operated in a sequential batch reactor (SBR) mode. All of the leachate generated by Column B was fed into a conditioning container, in which NH_4Cl or distilled water was added to change the NH_3–N concentration to 1500 ± 100 mg/L. Then the conditioned leachate (Stream IV, S4) was fed into the nitrifying reactor daily. The daily load of leachate to the nitrifying reactor was about 3.0 ± 0.5 L/d. The aeration rate was 0.375 m^3/h for a reactor volume of 0.009 m^3. Aeration continued for 23 h, followed by sludge settling for 30 min and supernatant discharge. The supernatant, whose volume was same as the leachate fed into the nitrifying reactor, was fed into the Column A, as S1. Because nitrifying bacteria are highly sensitive to pH and the rate of NH_3–N

Table 1
Characteristics of the synthetic refuse

Composition of the synthetic refuse							
Composition	Food waste	Plastic	Paper	Textile	Glass and metal	Total	Moisture content
Mass content (%)[a]	53	16	22	3	6	100	74.5
Elements in the synthetic refuse[a]							
Moisture content	Element composition						
	C	H	N	S	O	Ash	Cellulose/lignin
74.5	27.30	3.60	2.10	0.27	34.60	32.20	2.73

[a] All the indices, except for moisture content, were based on dry weight of the refuse.

oxidation reduces as the pH enters the acidic range. The pH value of the liquor in the SBR was adjusted by dosing it with sodium carbonate (Na_2CO_3, 1 mol/L. The added amount was equal to the mole amount of NH_3–N in the influent. Na_2CO_3 was separated to five shares equally, and added into the nitrifying reactor every 2 h with the peristalsis pump) on days 40 and 100, respectively, to investigate how pH affected the system performance. The testing period from days 1 to 40 was termed the "first stage" in this investigation, while the period from day 41 onwards was termed the "second stage".

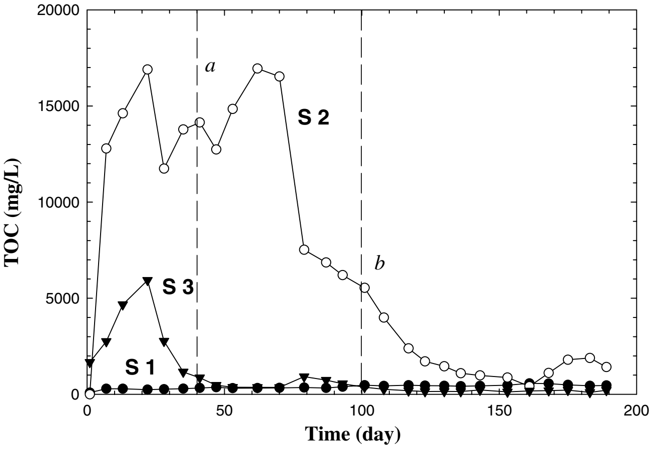
The chemical oxygen demand (COD), the conventional 5 days biochemical oxygen demand (BOD_5), total Kjeldahl nitrogen (TKN), NH_3–N, NO_2^-–N and NO_3^-–N of all liquor streams were measured according to the Standard Methods (APHA, 1998). The pH values were measured by use of a PHS-25 digital pH meter (Shanghai Precision & Scientific Instrument Co., Ltd.). Total organic carbon (TOC) and inorganic carbon (IC) were determined using the TN_b/TC multi N/C 3000 Analyzer (Analytik Jena AG). The furnace temperature was set to 850 °C, while the sample volume was 0.6 ml. The gas flow rate was measure by the wet gas flowmeter (Shanghai Gas Meter & Appliance Co., Ltd.) and the pressure difference of the gas in the effluent gas tube and the atmosphere was less than 0.001 atm. The gas was sampled on the gas effluent tubes with gas-tight micro syringes (Hamilton Ltd.). The methane and carbon dioxide content in biogas samples were analyzed using a gas chromatograph (GC102M, Shanghai Precision & Scientific Instrument Co., Ltd.) fitted with a thermal conductivity detector (TCD) and a 2-m stainless steel column packed with activated carbon (60/80 mesh). The temperatures of the injection port, oven, and TCD were 140, 120, and 140 °C, respectively. Helium was the carrier gas, and had a flow rate of 30 ml/min. The refuse sample, dried and ground first, was analyzed for its contents of carbon (C), hydrogen (H), nitrogen (N), and sulfur (S) using carbon, hydrogen, nitrogen, and sulfur determinator (CHNS-932, LECO Instruments Ltd.). High temperature combustion is used as the means of removing the elements from the solid sample. The sample is combusted in the heated oxygen-rich environment. The products of combustion are CO_2, H_2O, N_2, and SO_x. The gases, which are carried through the system by the helium carrier, are swept through the oxidation tube to complete the conversion to SO_2. The H_2O is swept through the non-dispersive infrared absorption detection system where it is measured. The N_2 is measured by TCD.

3. Results and discussion

3.1. First stage

Over the first 40 days of testing, the refuse hydrolyzed rapidly in Column A deduced from the rapidly accumulated TOC and NH_3–N in the leachate (S1 and S2 in Figs. 2 and 3). The pH of S2 was consistently below 6.0 (Fig. 4). Meanwhile, the corresponding NO_2^-–N and NO_3^-–N concentrations were also low, indicating that Column A could effectively remove nitrogen from the leachate stream (Fig. 5). Despite residual air being expelled during the first ten days, Gas A was composed mainly of CO_2 (Fig. 6), and was a product of refuse hydrolysis as well as NO_2^-–N/NO_3^-–N denitrification. The corresponding N_2 level was approximately 10%.

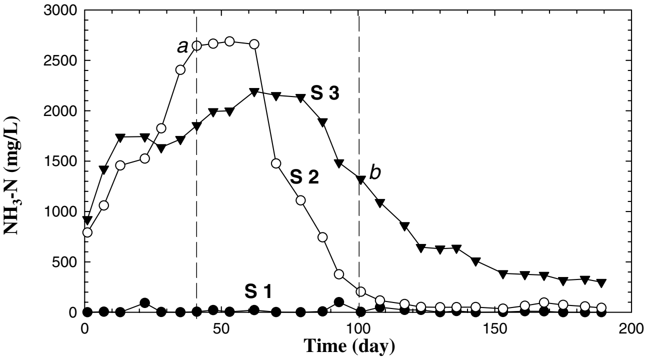
The high TOC and NH_3–N/low NO_2^-–N and NO_3^-–N stream from Column A was fed into Column B. It was noticed that in Column B markedly reduced the TOC level of leachate S2 (Fig. 2), raised the suspension pH to 8.0 (Fig. 4), owing to methanogenesis; and removed the resid-



a Na₂CO₃ was added to the nitrifying reactor (Days 40 to 99) to raise the pH of SI to >8.0.

b Na₂CO₃ was added to the nitrifying reactor (from Day 100) to raise the pH of SI to >9.0.

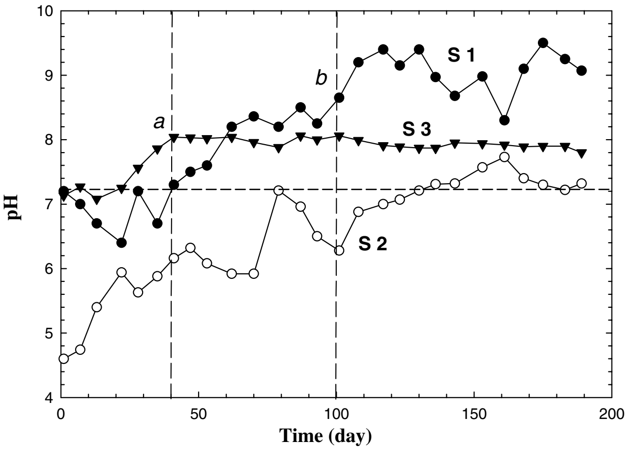
Fig. 2. Time evolution of total organic carbon (TOC) in leachates (S1, S2 and S3).



a Na$_2$CO$_3$ was added to the nitrifying reactor (Days 40 to 99) to raise the pH of SI to >8.0.

b Na$_2$CO$_3$ was added to the nitrifying reactor (from Day 100) to raise the pH of SI to >9.0.

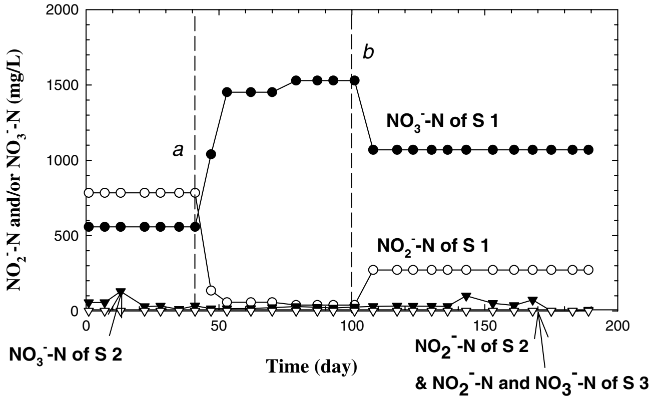
Fig. 3. Time evolution of NH_3–N in leachates (S1, S2 and S3).



a Na$_2$CO$_3$ was added to the nitrifying reactor (Days 40 to 99) to raise the pH of SI to >8.0.

b Na$_2$CO$_3$ was added to the nitrifying reactor (from Day 100) to raise the pH of SI to >9.0.

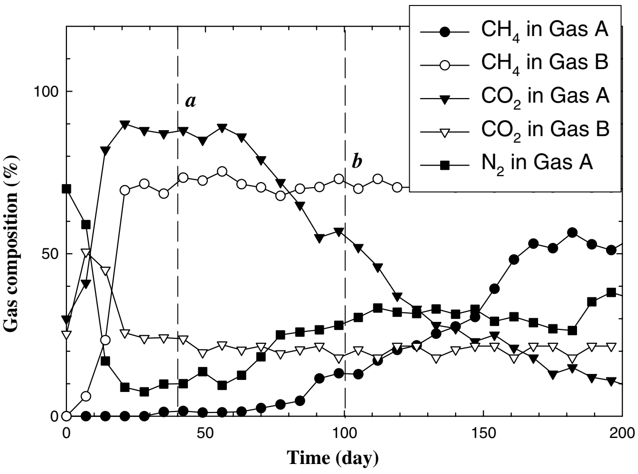
Fig. 4. Time evolution of pH in leachates(S1, S2 and S3).



a Na$_2$CO$_3$ was added to the nitrifying reactor (Days 40 to 99) to raise the pH of SI to >8.0.

b Na$_2$CO$_3$ was added to the nitrifying reactor (from Day 100) to raise the pH of SI to >9.0.

Fig. 5. Time evolution of NO_2^-–N and NO_3^-–N in leachates (S1, S2 and S3).



a Na$_2$CO$_3$ was added to the nitrifying reactor (Days 40 to 99) to raise the pH of SI to >8.0.

b Na$_2$CO$_3$ was added to the nitrifying reactor (from Day 100) to raise the pH of SI to >9.0.

Fig. 6. Time evolution of gas compositions in gas streams.

ual NO_2^-–N and NO_3^-–N in S2 (Fig. 5). The NH_3–N in S2 was somehow “trapped” by and later “released” from Column B to induce the delay of NH_3–N concentration in S3 (Fig. 3), probably owing to the retention of S2 in Column B for some time, the mixing of S2 with the liquid originally stayed in Column B, and the anisotropic liquid flow in Column B. The methane content of Gas B increased to approximately 70% from around day 20 (Fig. 6). The remaining gas was primarily CO_2. It was deduced that Column B removed TOC by converting it to methane and CO_2, with little NO_2^-–N and NO_3^-–N removal.

Subsequently, the low TOC/high NH_3–N stream from Column B was fed into the nitrifying reactor. The nitrifying reactor effectively converted the NH_3–N into NO_2^-–N and NO_3^-–N (Figs. 3 and 5), and further reduced residual TOC from Column B (Fig. 2), although such reduction was limited. The pH of S1 decreased to below 7.2, probably because hydrogen ions are yielded during NH_3–N oxidation. In addition, the concentration of NO_2^-–N exceeded that of NO_3^-–N in S1, probably because the specific growth rate of *Nitrobacter* is reduced when pH < 7.2 (Camilla and Gunnel, 2001).

3.2. Second stage

Na_2CO_3 salt was added to the nitrifying reactor from day 40 (point ‘a’ in Fig. 2) to control the pH of S1, which was influenced by S4, whose pH was similar as S3. The pH of S1 was raised to over 8.0 in about 20 days (Fig. 4), at which point the NH_3–N concentration in S2 was 2800 mg/L (Fig. 3). The change in pH facilitated the activity of NO_2^-–N oxidation bacteria in the nitrifying reactor to convert most NO_2^-–N (from 800 to nearly 0 mg/L) to NO_3^-–N (from 700 to 1500 mg/L) (S1 in Fig. 5). The pH of S2 fluctuated between 6 and 7 (Fig. 4). Moreover, the TOC of S2 started to decrease from day 70 since the hydrolysis of the refuse in Column A decreased and less organic carbon entered the leachate stream (Fig. 2).

During the second stage, the nitrogen content in Gas A gradually increased from 10% to 30% after day 75 (Fig. 6). Meanwhile, methanogenesis was partially established in

Column A, as demonstrated by the increasing CH_4 concentration in Gas A. Nevertheless, the change in the performance of Column A, which determined the characteristics of S2, did not significantly influence that of Column B. Owing to the "shielding" of Column B, the performance of the nitrifying reactor remained basically unchanged. The slow increase in the pH of the nitrifying reactor should be attributable to the dilution effects of alkaline S3 from Column B.

On day 100, the nitrifying reactor was dosed with extra Na_2CO_3 salt to increase the pH of S1 to above 9 (point 'b' in Fig. 4). This increase in pH partially depressed the activity of the nitrite-oxidizing bacteria in the nitrifying reactor, thus slightly increasing the NO_2^-–N concentration (S1 in Fig. 5). Additionally, the pH of S2 increased to >7.0 (Fig. 4). The efficiency of denitrification in Column A (S2 in Fig. 5) and methane production in Columns A and B (Gas A and Gas B in Fig. 6) remained little affected.

3.3. *Mass balance in different units*

The nitrogen and carbon balance was calculated as follows:

$$\text{Accumulated nitrogen/carbon species in liquid phase} = \sum C_i \times V_i,$$

where C_i is the concentration of the separate species (TKN, NO_3^-–N, NO_2^-–N, IC, or TOC) in the liquid phase in day *i*. V_i is the volume of the separate liquid (S1, S2 and S3) in day *i*.

The added amount of Na_2CO_3 in the nitrifying reactor was added into IC of S4. The amount of IC in S4 was equal to the measured IC in S4 and the carbon in the added Na_2CO_3.

$$\begin{aligned}\text{Accumulative nitrogen quantity converted into sludge} &= \text{accumulated TKN in influent}\\ &\quad - \text{accumulated TKN in outflow}\\ &\quad - \text{accumulated } NO_3^-\text{–N} - \text{accumulated } NO_2^-\text{–N},\end{aligned}$$

$$\text{Accumulated nitrogen in gas} = \sum \frac{p_{N_2,i} \times V_i}{22.4} \times 28,$$

$$\text{Accumulated C to methane} = \sum \frac{p_{CH_4,i} \times V_i}{22.4} \times 12,$$

$$\text{Accumulated C to carbon dioxide} = \sum \frac{p_{CO_2,i} \times V_i}{22.4} \times 12,$$

where $p_{N_2,i}$ is the volume fraction of the N_2 in the generated gas in day *i*. $p_{CH_4,i}$ is the volume fraction of the CH_4 in the generated gas in day *i*. $p_{CO_2,i}$ is the volume fraction of the CO_2 in the generated gas in day *i*. V_i is the volume of the generated gas in standard status in day *i*.

3.3.1. *Nitrifying reactor*

According to Table 2, over 90% of TKN in the influent was transformed into NO_2^-–N or NO_3^-–N, and less than 10% was possibly transformed into biomass. The accumulative flow of TKN into the nitrifying reactor was 844 g N on day 189, while the corresponding accumulative nitrogen content of NO_2^-–N and NO_3^-–N was 777 g N, representing 92% of the nitrogen conversion. Kinetic studies (Fig. 7) demonstrated that the nitrification followed zero-order kinetics, with a time constant of around 35 mg/L-h (data not shown). Consequently, assuming an influent that contains 500 mg/L NH_3–N, the nitrifying reactor would take approximately 14.5 h to fully oxidize the NH_3–N.

Table 2
Nitrogen balance in the nitrifying reactor

Time (d)	TKN in S4 (g N)	TKN in S1 (g N)	NO_3^-–N in S4[a] (g N)	NO_2^-–N in S4[a] (g N)	NO_3^-–N in S1 (g N)	NO_2^-–N in S1 (g N)	N in sludge (g N)
1	2.3	0.0	–	–	0.0	2.0	0.3
7	15.8	0.2	–	–	2.1	12.6	0.9
28	109.2	1.8	–	–	30.0	67.1	10.2
63	291.9	2.7	–	–	183.3	73.6	32.3
91	397.4	3.0	–	–	295.8	81.7	16.9
119	529.7	5.5	–	–	338.4	127.8	58.0
154	687.2	6.2	–	–	374.8	250.5	55.7
182	813.2	6.6	–	–	360.5	371.4	74.7
189	844.7	6.6	–	–	365.5	411.6	61.0

[a] The values were nearly 0.

Table 3 shows the carbon balance for the nitrifying reactor. The TOC reduction was approximately 10%, primarily converted into biomass and CO_2. For example, on day 189 the accumulative TOC amount from the conditioned stream (S4) was 268.8 g C, while that for discharged leachate (S1) was 228.5 g C. This low TOC reduction was partially attributable to the low BOD_5/COD ratio for the leachate (<0.1). Meanwhile, the relatively high IC content from Column B was used to neutralize the H^+, produced from nitrification, and possibly to convert to CO_2 and release to the atmosphere.

3.3.2. *Column A*

Table 4 shows the nitrogen balance in Column A. The nitrogen originally residing of the MSW in the column totaled approximately 5 g N, which was gradually expelled

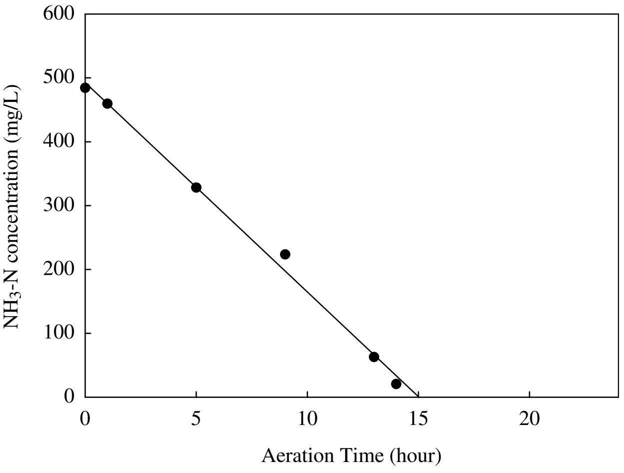


Fig. 7. Time evolution of NH_3–N concentration in the nitrifying reactor.

Table 3
Carbon balance in the nitrifying reactor

Time (d)	TOC in S4 (g C)	IC in S4 (g C)	TOC in S1 (g C)	IC in S1 (g C)
1	1.2	9.1	0.0	0.1
7	8.4	57.7	6.7	0.5
28	33.6	193.3	28.9	1.7
63	75.6	378.3	69.6	3.8
91	109.2	517.4	96.1	5.5
119	151.2	632.3	134.6	7.2
154	203.7	781.8	187.4	9.3
182	258.3	899.4	224.7	11.0
189	268.8	928.8	228.5	11.4

during the first 10 days, as indicated by Gas A on day 1 in Table 4. During the test most NO_2^-–N/NO_3^-–N fed into the column was converted into nitrogen gas. For example, on day 210, the accumulative nitrogen production was 125.3 g N (130.3–5), while the total consumption of NO_2^-–N and NO_3^-–N was 114.1 g N (86.7 g N for NO_3^-–N and 27.4 g N for NO_2^-–N). Meanwhile, the net production of N from the refuse was 97.6 g N on day 210, produced through refuse hydrolysis. Notably, the accumulative nitrogen amount (125.3 g N) was higher than that of the hydrolyzed amount (97.6 g N), which was attributed to the addition of NH_4Cl to the conditioning container.

Until day 210, the accumulative carbon released from the refuse was 895.3 g C, 5.0% of which was converted into inorganic carbon in leachate, with 72.1% being converted into TOC in leachate, 24.4% being converted into landfill gas, 20.9% to CO_2 and 3.5% to CH_4 (Table 5).

3.3.3. Column B

As illustrated in Table 6, a time lag existed in Column B between the TKN contents in the recycled and in discharged leachate, probably owing to the mixing of S2 and the liquid in Column B and the anisotropic flow in the column. This enabled the well-decomposed layer to act not only as a methane producer, but also to provide the system with a TKN shock load buffer. The accumulative discharged TKN became close to the recycled TKN following day 190. Consequently, all delayed nitrogen would eventually be released back to the leachate. Column B did not produce detectable levels of nitrogen gas.

On day 200, the influent TOC was converted to carbonate (44%), CO_2 gas (14%) and CH_4 (29%), while 13% TOC remained in the leachate (Table 7). Sludge production was negligible. The high inorganic carbon content (IC > 3000 mg/L) was partially attributable to the dissolution of CO_2 in alkaline leachate (pH > 7.8).

3.4. Discussion

Following 200 days of treatment, the refuse in Column A was stabilized (i.e., the nitrogen-containing substances were converted and removed and the ratio of cellulose/lignin was below 0.1). At the end of the test, the mass (dry weight) of the refuse was reduced by 66%.

Table 4
Nitrogen balance in Column A

Time (d)	TKN in S1[a] (g N)	TKN in S2 (g N)	N in gas (g N)	NO_3^-–N in S1 (g N)	NO_3^-–N in S2 (g N)	NO_2^-–N in S1 (g N)	NO_2^-–N in S2[a] (g N)	N in refuse[b] (g N)
1	–	2.0	0.0	0.6	0.0	0.8	–	111.9
28	–	32.4	28.0	7.3	0.8	10.2	–	–
63	–	72.3	45.1	19.2	1.1	16.3	–	–
91	–	92.1	66.7	41.6	1.4	16.9	–	–
119	–	96.2	87.7	57.7	1.8	19.8	–	–
154	–	97.8	112.6	75.1	2.7	24.2	–	–
182	–	99.1	125.5	83.4	2.8	26.4	–	–
210	–	99.7	130.3	89.5	2.8	27.4	–	14.3

[a] The values were nearly 0.

[b] Measured by CHNS-932.

Table 5
Carbon balance in Column A

Time (d)	TOC in S1 (g C)	IC in S1 (g C)	TOC in S2 (g C)	IC in S2 (gC)	C to CH_4 (g C)	C to CO_2 (g C)	C in landfilled waste[a] (g C)
1	0.0	0.1	20.0	0.0	0.0	1.6	1142.3
28	0.5	0.5	252.0	0.8	0.0	77.7	–
63	1.0	1.0	459.9	3.4	0.9	135.6	–
91	1.5	1.7	563.6	8.9	3.3	159.4	–
119	2.0	2.5	614.8	18.0	7.8	174.2	–
154	2.2	3.8	632.2	36.5	17.6	183.3	–
182	2.5	4.6	643.6	45.1	27.7	186.7	–
210	2.7	5.5	648.5	50.0	31.2	187.3	247.0

[a] Measured by CHNS-932.

Table 6
Nitrogen balance in Column B

Time (d)	TKN in S2 (g N)	TKN in S3 (g N)	NO_3^-–N in S2 (g N)	NO_3^-–N in S3 (g N)	NO_2^-–N in S2[a] (g N)	NO_2^-–N in S3[a] (g N)	N in gas (g N)	N in refuse[b] (g N)
1	2.0	0.0	0.0	0.0	–	–	15.6	2.0
28	32.4	20.8	0.8	0.1	–	–	28.4	–
63	72.3	46.5	1.0	0.2	–	–	34.2	–
91	92.1	68.3	1.3	0.2	–	–	35.4	–
119	96.2	83.3	1.7	0.3	–	–	36.5	–
154	97.8	92.0	2.5	0.3	–	–	36.8	–
182	99.1	94.4	2.7	0.3	–	–	37.0	–
210	99.7	96.0	2.7	0.3	–	–	38.2	6.1

[a] The values were nearly 0.

[b] Measured by CHNS-932.

Table 7
Carbon balance in Column B

Time (d)	TOC in S2 (g C)	IC in S2 (g C)	TOC in S3 (g C)	IC in S3 (g C)	C to CH_4 (g C)	C to CO_2 (g C)
1	3.0	0.0	0.0	2.7	0.0	0.0
28	184.9	0.8	51.1	67.8	46.6	39.2
63	382.1	3.4	61.5	163.7	122.8	63.2
91	463.4	18.2	69.3	215.7	137.7	67.6
119	507.4	50.8	72.6	271.9	150.9	71.1
154	521.1	78.3	75.2	309.1	153.8	72.0
182	529.9	89.4	76.4	323.5	156.4	72.7
210	533.8	95.6	77.8	330.6	157.9	73.2

Using this process, managing the nitrogen in leachate generated from 1 kg of refuse (dry basis) would produce 18.45 g N into N_2 gas, and convert 24.9 g C to CH_4 and 86.7 g C into CO_2. This would account for 86% and 42% of the original nitrogen and carbon contents in the original refuse, respectively. TOC removal was also assisted by the conversion of TOC into IC in the leachate.

It is clear that balanced functioning of all three units mentioned above is required to achieve good nitrogen removal. When S3 was characterized as low TOC and low nitrogen species concentration, the stream could be discharged without the treatment by the nitrifying reactor. However, if Column B had not existed and the leachate generated by Column A (S1) had been directly recycled to the nitrifying reactor, in the initial stage high content of TOC in S1 (because of the rapid hydrolysis of the fresh MSW in Column A) would stimulate the growth of heterotrophic bacteria in the nitrifying reactor and inhibit the growth of nitrifying bacteria in the nitrifying reactor. On the other hand, if Column B were not functioning properly owing to the strongly acidic leachate fed by Column A (itself probably due to the rapid hydrolysis reaction or to the possible inhibition effects by the high volatile fatty acid level), the methane yielded in Column B would decline and the alkaline produced by methanogenesis would become insufficient to buffer the acids in S3. The TOC concentration in S1 then would be high, thereby also facilitating the conversion of nitrogen to biomass rather than to NO_2^-–N/NO_3^-–N. These occurrences would inevitably produce sludge disposal problems.

After the operation for about 200 days, the COD concentration of S3 decreased to less than 300 mg/L, and has met the second level of the current discharge standards in China (<300 mg/L, GB16889-1997, State Environmental Protection Agency of PR China, 1997). NH_3–N concentration was approximately 230 mg/L, which was higher than the discharge standard mentioned above (<25 mg/L). However, the NH_3–N concentration in S2, as the source of the NH_3–N in S3, has decreased to 43 mg/L. The high NH_3–N concentration of S3 was attributed to the NH_3–N retained in the Column B. It could be deduced that the NH_3–N concentration of S3 would decrease lower than the discharge standard as long as the operation continues for a long time. This means the nitrogen management scheme recommended in this paper could possibly satisfy the requirement of the pollution control of the landfill. However, for the application of the scheme in the real case, several design and operational approaches have to be abided by. First of all, the landfill should be divided into several cells, in which the leachate could be collected. Then, when the landfill operation starts in one cell, a thick layer of well-decomposed MSW should be placed at the bottom of the cell, acting as the methanogenesis area. Above the well-decomposed MSW layer, the fresh refuse could be landfilled continuously. The leachate generated by the fresh MSW layer first leaches though the methanogenesis area and the TOC is removed. Then, the leachate is collected and transferred to the ex situ biological nitrifying reactor, in which the NH_3–N is converted to NO_3^-–N and NO_2^-–N. The nitrified leachate is then recycled into the fresh waste layer to accelerate the waste stabilization. When the landfilled MSW in the cell reaches a certain quantity (F), the landfill process stops and the final cover system is constructed, the nitrifying and recycling process continues. At the same time, another cell starts the same landfilling and leachate treatment/recycling operation. The number of such cells (N) necessary for the landfill site depends on the time scale (t) during which the leachate generated by a cell is completely treated and reaches the discharge standard. Then, the leachate of the cell would be discharged and a new landfilling process would start based on this cell. The relationship among the parameters, F, N, and t, could be demonstrated with the equation as follows:

$$N = \frac{W}{F} \cdot t,$$

where W is the total amount of the MSW treated by the landfill site.

A separate conditioning pond should be equipped for every landfill cell. The conditioning capacity should be more than the total volume of the leachate generated in t days.

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References

APHA, AWWA, WEF, 1998. Standard Methods for the Examining of Waste and Wastewater, 20th ed. American Public Health Association, Washington, DC.

Barlaz, M., Reinhart, D., 2003. Editorial. Waste Management 23, 557–559.

Berge, N.D., Reinhart, D.R., 2003. In-situ nitrification of leachate in bioreactor landfills. In: Proceedings of Sardinia 2003 Ninth International Waste Management and Landfill Symposium, Cagliari, Italy.

Burton, S.A.Q., Watson-Craik, I.A., 1999. Accelerated landfill refuse decomposition by recirculation of nitrified leachate. In: Proceedings of Sardinia 99 Seventh International Waste Management and Landfill Symposium, Cagliari, Italy.

Camilla, G., Gunnel, D., 2001. Development of nitrification inhibition assays using pure cultures of nitrosomonas and nitrobacter. Water Research 35, 433–440.

He, P.J., 2002. The research on the feasibility of the 4th stage project of the Laogang MSW Landfill in Shanghai, PR China. Report to Shanghai City Appearance And Environmental Sanitary Administrative Bureau. Shanghai City Appearance And Environmental Sanitary Administrative Bureau, Shanghai, PR China.

He, P.J., Shao, L.M., Qu, X., Li, G.J., Lee, D.J., 2005. Effects of feed solutions on refuse hydrolysis and landfill leachate characteristics. Chemosphere 59, 837–844.

Jokela, J.P.Y., Kettunen, R.H., Sormunen, K.M., Rintala, J.A., 2002. Biological nitrogen removal from municipal landfill leachate: low-cost nitrification in biofilters and laboratory scale in-situ denitrification. Water Research 36, 4079–4087.

Keenan, J.D., Steiner, R.L., Fungaroli, A.A., 1984. Landfill leachate treatment. Journal of the Water Pollution Control Federation 56, 27–33.

Mehta, R., Barlaz, M.A., Yazdani, R., Augenstein, D., Bryars, M., Sinderson, L., 2002. Refuse decomposition in the presence and absence of leachate recirculation. Journal of Environmental Engineering 128, 228–236.

Onay, T.T., Pohland, F.G., 1998. In situ nitrogen management in controlled bioreactor landfills. Water Research 32, 1382–1392.

Pohland, F.G., 1995. Landfill bioreactor: historical perspective, fundamental principles, and new horizons in design and operations. In: Landfill Bioreactor Design and Operation Sem. Proc., EPA/600/R-95/146, pp. 9–24.

Price, G.A., Barlaz, M.A., Hater, G.R., 2003. Nitrogen management in bioreactor landfills. Waste Management 23, 675–688.

Reinhart, D.R., McCreanor, P.T., Townsend, T., 2002. The bioreactor landfill: its status and future. Waste Management and Research 20, 172–186.

State Environmental Protection Agency of PR China, 1997. Standard for pollution control in municipal solid waste landfill. GB16889-1997.