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Alteration of moving bed sequencing batch reactor operational strategies for the enhancement of nitrogen removal from stabilized landfill leachate

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ABSTRACT

Stabilized landfill leachate is well known to contain high concentration of nitrogen particularly in the ammonium nitrogen (NH_4^+-N) form. Also considering of high toxicity of non-biodegradable and low proportion of biodegradable compounds in stabilized landfill leachate, the conventional biological treatment of nitrogen is usually inefficient. As such, the quest of this study was to revise the operational strategies of bioreactor, i.e. moving bed sequencing batch reactor (MBSBR), for the enhancement of nitrogen removal from stabilized landfill leachate. The performance of MBSBRs packed with 8% (v/v) of polyurethane (PU) cubes and polyethylene (PE) rings, respectively, in removing nitrogen from a combined stabilized landfill leachate and domestic wastewater in increasing leachate volumetric ratio under various operational strategies was investigated. On increasing the leachate volumetric ratio to 14%, the performance of both MBSBRs in NH_4^+-N removal was comparable under the continuous aeration strategy. When the aeration strategy was changed to intermittent aeration (IA), the MBSBR with PU media achieved a higher NH_4^+-N removal rate, while the MBSBR with PE media failed to remove NH_4^+-N completely. For the MBSBR with PU media, the adoption of IA coupled with step feeding strategy had yielded a removal efficiency of 80% for total nitrogen in comparison to 64% if only IA strategy was adopted at the leachate volumetric ratio of 14%. Nevertheless, further increase in leachate volumetric ratio to 20% had led to the accumulation of NH_4^+-N in the effluent of MBSBR under IA-SF strategy. At this stage, the MBSBR operated with IA strategy could still remove NH_4^+-N completely. Thus, the IA-MBSBR with PU media is the preferred operational strategy for nitrogen removal at higher leachate volumetric ratio.

Keywords: Nitrogen removal; Stabilized landfill leachate; Moving bed sequencing batch reactor; Support media; Aeration strategy Step feeding

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1. Introduction

For decades, sanitary landfilling has become the most widely employed method for the disposal of solid wastes. Accordingly, up to 95% of municipal solid waste collected is disposed of in more than 150,000 landfills worldwide [1,2]. Nonetheless, one of the major environmental problems associated with landfilling application is the generation of large quantity of leachate, hazardous, and heavily polluting wastewater, which has the potential of causing severe water pollution if not properly treated. Hence, the management of landfill leachate is recognized as a critical issue interlocked with environmental processing of sanitary landfills [3–5].

Leachates from stabilized landfills which have been operated for more than 10 years [6] contain large amounts of non-biodegradable organic substances such as fulvic and humic compounds with a chemical oxygen demand (COD) range of 500–4,500 mg L^{-1} and high ammonium nitrogen (NH_4^+-N) concentration of more than 400 mg L^{-1} [5]. Treatment approaches including the physical methods of ammonia stripping, floatation, and activated carbon adsorption and the chemical methods of coagulation–flocculation, Fenton reagents, and ozone oxidation have been well exploited by researchers over the years [7–12]. Many of these methods, however, are very expensive due to the requirement of large amount of chemical dosing and electricity to yield high-quality treated leachate. Based on economic and environmental considerations, biological process seems to be the leading alternative for the treatment of stabilized landfill leachate. For instance, in nitrogen removal, the biological nitrification and denitrification reactions can transform NH_4^+-N into nitrogen gas (N_2) without secondary pollution [12–15]. However, the removal of nitrogen from stabilized landfill leachate without the addition of external carbon source during denitrification always poses a problem because leachate contains very low amount of easily biodegradable carbon source. Thus, recent research interest has been directed towards the modification of conventional bioreactors for the enhancement of biological nitrogen removal.

Of late, the use of moving bed sequencing batch reactor (MBSBR) which combines suspended growth and attached growth processes in a single reactor to remediate nitrogen bearing wastewater has gained increasing interest among the researchers [16–20]. In general, the commercially available media, namely Kaldnes (polyethylene material) and Linpor (high porosity plastic material) are used as support media in MBSBR due to their ability to suspend in mixed liquor. Besides, both media have been proven to possess specific advantage in promoting biological nitrogen removal, e.g. high external surface area of Kaldnes media can enhance nitrification process [21] and high porosity of Linpor media can spur denitrification process [22,23]. Thus, the selection of support media in MBSBR is highly consequential in maximizing nitrogen removal from stabilized landfill leachate.

For biological nitrogen removal, the simultaneous nitrification and denitrification (SND) process is considered as the most cost-effective path for biological nitrogen removal due to the requirements of low-dissolved oxygen concentration and external carbon source in addition to time saving [21,24]. As such, the operational strategies of MBSBR, particularly the aeration strategy and influent feeding mode must be carefully altered and fine-tuned to ensure SND is attained in the bioreactor. In studying the aeration strategy, Lim et al. [21] reported that higher nitrogen removal efficiency via SND process could be gained by converting continuous aeration (CA) to intermittent aeration (IA) strategies in MBSBR. With regard to influent feeding, Puig et al. [25] reported that incorporating step feeding (SF) mode during the anoxic phase of six anoxic-aerobic events could lead to near complete nitrogen removal. The total nitrogen (TN) removal efficiency was also found to increase with increasing feeding steps [26]. To date, the strategy involving IA coupled with SF was reported to apply mainly on the treatment of domestic and municipal wastewaters [27–30], but seldom used for leachate treatment. It is therefore of great research interest to explore the potential of this strategy in promoting SND process in the treatment of stabilized landfill leachate.

The co-treatment of landfill leachate and domestic wastewater is essential due to the toxicity of the leachate. Nonetheless, only co-treatment studies involving low volumetric ratio of leachate had been reported [31,32]. Presently, reports on the feasibility of using MBSBR in enhancing nitrogen removal from a combined landfill leachate and domestic wastewater are also lacking. Therefore, the first objective of this research was to evaluate the performance of MBSBRs packed with polyethylene (PE) cylindrical rings (Kaldnes media) and polyurethane (PU) foam cubes (Linpor media), respectively, in the removal of nitrogen from combined stabilized landfill leachate and domestic wastewater. The second objective was to ascertain the most appropriate aeration strategy and influent feeding mode based on the highest TN removal at increasing volumetric ratio of stabilized landfill leachate in the combined wastewater.

2. Materials and methods

2.1. Leachate sampling site and characteristics

The raw leachate was collected from the Pulau Burung landfill site situated within the Byram Forest Reserve in Penang, Malaysia. This landfill site was initially operated as a semi-aerobic system complying with the Level II sanitary landfill standards, but was later upgraded to Level III by employing controlled tipping with leachate recirculation. The generated leachate is collected and flows into detention ponds. In this study, leachate samples were collected from one of the ponds and swiftly transported to the laboratory. The fresh samples were analyzed for its characteristics in accordance with the Standard Methods [33] and the results are shown in Table 1. The remaining leachate samples were kept at 4°C prior to use. Several researchers had reported low BOD_5/COD ratio and high NH_4^+-N concentration for the leachate from this landfill signifying high stability and low biodegradability [34].

2.2. Experimental setup and operation

Three identical plexiglass reactors with the dimension of 25 cm × 25 cm × 40 cm (Length × Width × Height) and working volume of 10 L were inoculated with activated sludge from a local sewage treatment plant and fed with simulated domestic wastewater containing (in mg L^{-1}) peptone (100), sucrose (280), CH_3COONa (600), $(NH_4)_2SO_4$ (250), KH_2PO_4 (100), K_2HPO_4 (100), $MgSO_4$ (49), $CaCl_2$ (30), and $FeCl_3{\cdot}6H_2O$ (10), giving rise to COD and NH_4^+-N concentrations of 800 and 50 mg L^{-1}, respectively, in influent. These compositions were previously mixed with tap water to acquire the elements of micronutrients required for the growth of micro-organisms in the activated sludge. The reactors were operated as sequencing batch reactors (SBRs) with an exchange volume of 70% and a cycle time of 24 h following the sequencing periods of FILL, REACT (aerobic + anoxic), SETTLE, DRAW, and IDLE at the time ratios of 2:12 (9 + 3):1.5:1:7.5. The mixed liquor was homogenized by an ejector throughout the REACT period. The aeration during the aerobic REACT period was provided by aquarium air pumps with measured dissolved oxygen concentration being maintained above 4.0 mg L^{-1}. The pH was well buffered around 7.8 ± 0.3 during the REACT period predominantly by the acetate and phosphates in the influent. Sufficient ethanol solution (theoretical COD of 75 mg O_2 mL^{-1}) with the selected ratio of COD/NO_3^--N (g/g) of approximately 7.0 was spiked into the reactors as an external carbon source at the beginning of the anoxic REACT period to reduce the remaining oxidized nitrogen in the mixed liquor to N_2. The sludge age was controlled at 30 d throughout the operational period.

Table 1
Characteristics of raw landfill leachate

Parameter[a]	Range value	Mean value
pH	8.3–8.5	8.4
NH_4^+-N	503–1,671	1,087
PO_4^{3-}	2.3–7.3	4.8
Cl^-	2,949–3,233	3,091
COD	2,056–2,960	2,508
BOD_5	23–54	39
Total solids	5,390–9,026	7,208
Heavy metals[b]	≤0.3	–

[a] Unit in mg L^{-1} except pH.

[b] Heavy metals determined were Cd, Cu, Ni, Pb, and Zn.

Once the activated sludge had acclimated to the simulated domestic wastewater after three months of operation, two of the SBRs were converted to MBSBRs by packing the working volume with 8% (v/v) of support media. Higher percentage of packing volume was avoided to ensure smooth circulation of support media in the mixed liquor. The MBSBRs with PU foam cubes and PE cylindrical rings as the support media were labeled as R-PU and R-PE, respectively. The characteristics of the two support media are shown in Table 2. The third reactor continued to be operated as a SBR and served as a control system (R-C). The operation of the three reactors was continued until the pseudo-steady state (defined as complete removal of NH_4^+-N within 5% variation of total reaction time required) was attained. At this state, the average mixed liquor volatile suspended solids concentration for all reactors was found to be in 2,500–3,000 mg L^{-1} and fluctuated within this range for all studies.

2.3. Operational strategies of MBSBRs for nitrogen removal from combined wastewater at various mixing volume ratios

Table 3 shows the experimental scheme involving various Phases of study. In Phases I–III, the reactors, namely R-C, R-PU, and R-PE were operated under CA strategy and fed with increasing NH_4^+-N concentration (due to increasing volumetric ratio of leachate) in the combined wastewater. From Phase IV onwards, the aeration strategy was converted to IA throughout the REACT period. The adopted IA strategy involved 1-h period of aeration followed by 1-h period of non-aeration and the pattern was repeated; thus, amounting to six aeration and non-aeration events. Also, starting

Table 2
Characteristics of support media

Support media[a]	PU foam cube	PE cylindrical ring
Dimensions (cm)	2 × 2 × 2 (Length × Width × Height)	0.64 × 0.43 × 1.00 (Outer diameter × Inner diameter × Length)
Density (kg m^{-3})	89 ± 6	95 ± 2
Volume of one support media (cm^3)	8	0.18
Quantity in MBSBR	100	4,444
Estimated surface area of one support media (cm^2)	47[b]	3.71
Estimated total surface area in MBSBR (m^2)	0.47	1.65

[a] Support media occupied 8% (v/v) of MBSBR's working volume.
[b] Based on estimates from Lim et al. [23].

Table 3
Experimental scheme for various operational strategies

Phase	Volumetric ratio of leachate in influent (%)	Influent NH_4^+-N concentration (mg L^{-1})	Aeration strategy[a]	Investigated reactor
I	2.4	75	CA	R-C, R-PU and R-PE
II	4.5	100	CA	R-C, R-PU and R-PE
III	14.3	200	CA	R-C, R-PU and R-PE
IV	14.3	200	IA	R-PU and R-PE
V	14.3	200	IA	R-IA and R-IASF
VI	20.0	250	IA	R-IA and R-IASF
VII	25.0	300	IA	R-IA

[a] CA: Continuous aeration; IA: Intermittent aeration.

from Phase IV, sufficient ethanol solution was spiked into the reactors at the beginning of the anoxic period of the sixth event. In Phase IV, the performance of R-PU and R-PE as MBSBR in nitrogen removal was then evaluated to select a better support medium used in later experiments.

Based on the results from Phase IV study which indicated that R-PU outperformed R-PE, the content of R-PU was used to setup two MBSBRs, each with a working volume of 3.3 L. The performance of the MBBSRs operated solely with IA strategy (R-IA) and with IA coupled with step feeding (R-IASF) in nitrogen removal at increasing volumetric ratio of leachate was evaluated in Phases V and VI. For R-IASF, the influent volume was divided equally into six portions with the first portion being added instantaneously to the reactor during the FILL period and the other portions added instantaneously during subsequent five anoxic periods. For both MBSBRs, sufficient ethanol solution was added at the start of sixth anoxic period to eliminate the residual oxidized nitrogen.

For each Phase, data collection were done during the profile studies of the REACT period in which nitrogen species concentrations, namely NH_4^+-N, NO_2^--N, and NO_3^--N, were determined by following the Standard Methods [33].

3. Results and discussion

3.1. Phases I–III studies: removal of nitrogen at increasing volumetric ratio of leachate

Figs. 1(a)–(c) and 2(a)–(c) show the NH_4^+-N, NO_2^--N, and NO_3^--N concentration profiles during the REACT (aerobic) and REACT (anoxic) periods in Phases I and III for reactors R-C, R-PU, and R-PE, respectively. It was evident that complete NH_4^+-N removal was achieved in all the reactors in Phase I, but only MBSBRs were able to remove NH_4^+-N completely in Phase III. The experimental NH_4^+-N concentration data for the period during which the formation of oxidized nitrogen was observed were found to be well fitted ($R^2 > 0.98$) to the pseudo-zero-order kinetic model and the mean pseudo-zero-order rate constants, k_{AN}, for NH_4^+-N removal in Phases I–III are shown in Table 4. The values of k_{AN} were found to increase

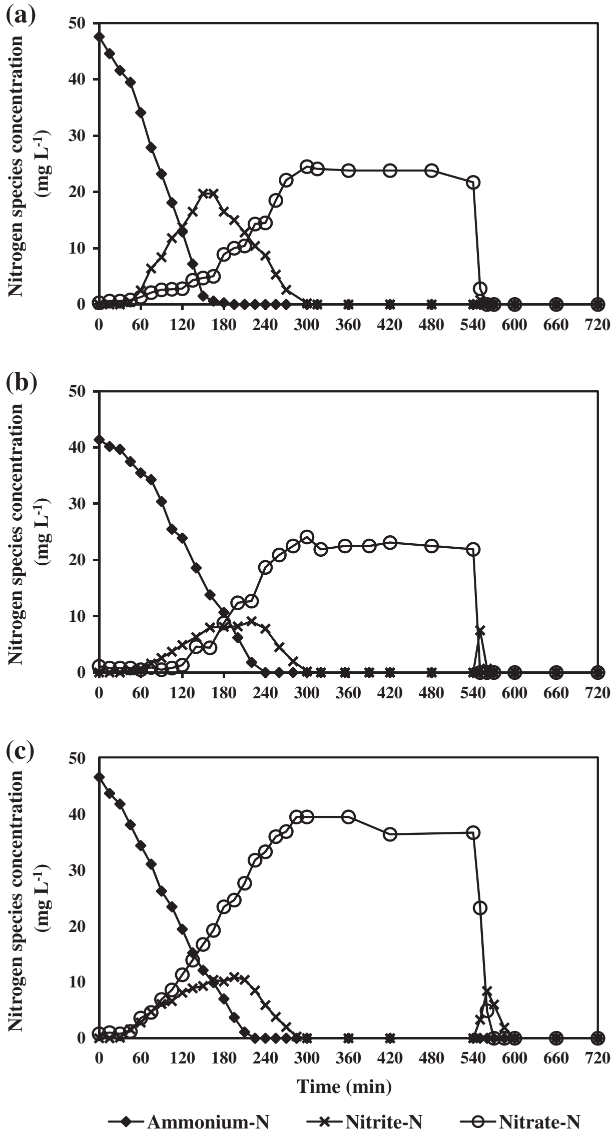

Fig. 1. Concentration profiles of nitrogen species during the REACT period for reactors (a) R-C, (b) R-PU, and (c) R-PE operated at leachate volumetric ratio of 2.4% and with CA strategy.

from Phases I to II for all reactors indicating an inducement of nitrification process when the leachate volumetric ratio was increased slightly from 2 to 5%. In a study on the co-treatment of leachate, Fudala-Ksiazek et al. [31] reported that the ammonia utilization rate rose with the increase in leachate dosage up to 5%, a result in agreement with this study. Faster nitrification rate in Phase II as compared to Phase I could plausibly be explained by the increase in influent NH_4^+-N concentration resulting in the proliferation of nitrifying bacteria. It was also observed that the k_{AN} values of R-C were higher than those of R-PU

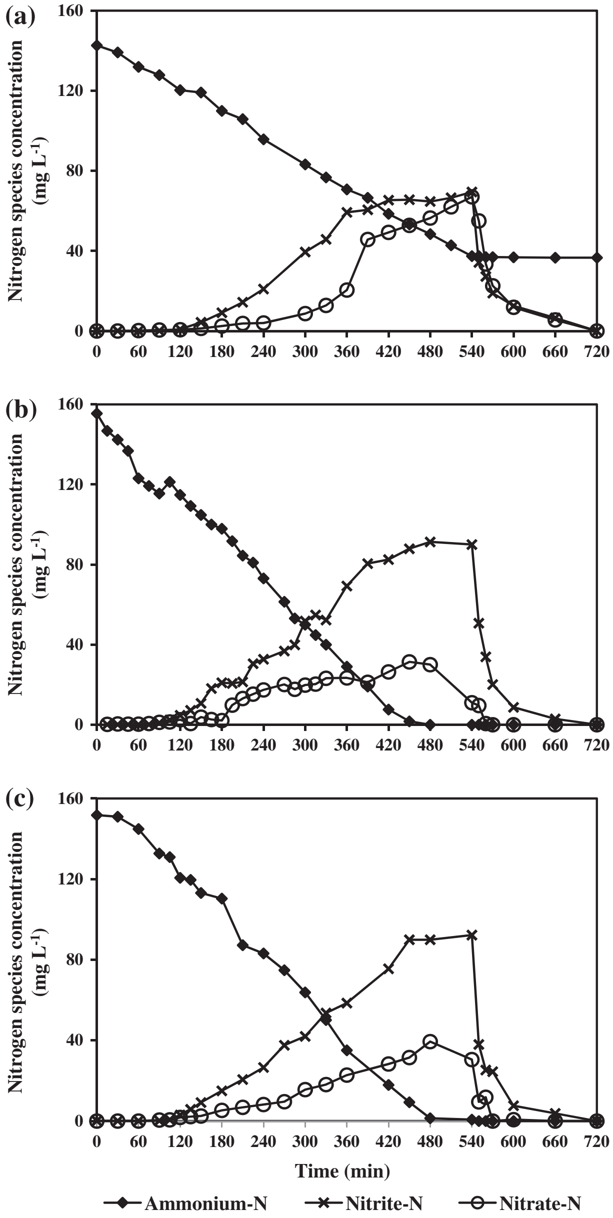

Fig. 2. Concentration profiles of nitrogen species during the REACT period for reactors (a) R-C, (b) R-PU, and (c) R-PE operated at leachate volumetric ratio of 14.3% and with CA strategy.

and R-PE in Phases I and II due to greater amount of suspended biomass in SBR relative to MBSBRs since 8% (v/v) of the working volume of MBSBRs was occupied by the support media. Although, the attached-growth biomass was also present in MBSBRs, but the mobility of attached-growth biomass was more restricted in comparison with the suspended-growth biomass. In any case, based on the finding of an earlier study [23], the attached-growth biomass on the

Table 4
Mean values of pseudo-zero-order rate constant of NH_4^+-N removal, k_{AN}, for various reactors in phases I–III

	k_{AN} (mg g^{-1} h^{-1})		
Phase	R-C	R-PU	R-PE
I	6.7 ± 1.1	5.9 ± 1.2	5.3 ± 0.4
II	11.6 ± 2.0	10.4 ± 1.4	7.9 ± 0.2
III	3.1 ± 0.4	7.1 ± 0.7	6.2 ± 1.3

external surface of the PU support media would have contributed only about 10% to the total biomass in the MBSBR reactor. Thus, in the absence of inhibitory effect, the probability of contact between the biomass and substrate becomes the dominant factor in the removal rate of NH_4^+-N due to nitrification process.

However, when the leachate volumetric ratio was increased to 14% in Phase III during which the influent NH_4^+-N concentration was increased to 200 mg L^{-1}, the rate of NH_4^+-N removal deteriorated in all the reactors. A drastic decrease in k_{AN} value from 11.6 to 3.1 mg g^{-1} h^{-1} was observed in the SBR in Phase III which explained the incomplete removal of NH_4^+-N (Fig. 2(a)). In case of MBSBRs, both reactors also registered a decrease in k_{AN} value to about 6.6 mg g^{-1} h^{-1} for both reactors. Several researchers had reported that the biological processes were more susceptible to the toxicity effect of high NH_4^+-N concentration when the volumetric ratio of leachate to domestic wastewater exceeded 10% [6,32,35]. As a whole, the MBSBRs were clearly more superior over the SBR system at higher stabilized leachate loading volume primarily due to the presence of attached-growth biomass on the support media surfaces. This type of biomass is typically less vulnerable to the toxicity effect of pollutants, which is normally deleterious to the suspended-growth biomass [36–38].

3.2. Phase IV study: selection of support media for MBSBR operated with IA strategy

The removal of nitrogen species during the REACT period in the MBSBRs packed with different support media and operated with IA strategy in Phase IV is shown in Fig. 3. It was observed that the rate of nitrification in reactor R-PU operated with IA strategy was enhanced with the period required for the complete removal of NH_4^+-N in the mixed liquor reduced to 4 h (Fig. 3(a)). The value of k_{AN} computed during the aeration period of R-PU in Phase IV was found to be 15.3 mg g^{-1} h^{-1}, which was doubled the value of the rate constant, i.e. 7.1 mg g^{-1} h^{-1}, attained in Phase III with CA strategy. In terms of aeration time for complete NH_4^+-N removal, a total period of only 4 h was required under the IA strategy as compared with 8 h under the CA strategy. In contrast, reactor R-PE failed to completely remove NH_4^+-N due to the inhibitory effect of leachate at the volumetric ratio of 14% when the aeration strategy was changed from CA to IA during the REACT period (Fig. 3(b)). The superiority of reactor R-PU to function under the IA condition without experiencing the conspicuous inhibitory effect of stabilized leachate in Phase IV as compared with R-PE could be explained by the presence of multilayers of PU foam cubes used as support media. The biomass grown in every layer was generally exposed to different levels of toxicity exerted by stabilized leachate with the outermost layer experiencing the highest toxicity level resulting in the greatest inhibition. Villaverde and Fernandez-Polanco [39] reported that the accumulation of inactive cells in the outer layer would provide the diffusion resistance to toxicity. Although the amount of attached-growth biomass on the external surface of the PU cubes only constituted

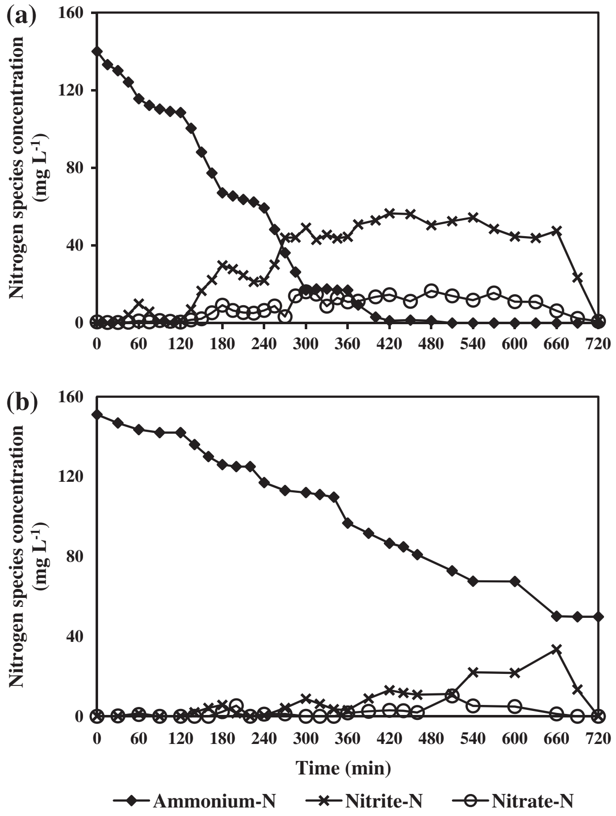

Fig. 3. Concentration profiles of nitrogen species during the REACT period for reactors (a) R-PU and (b) R-PE operated at leachate volumetric ratio of 14.3% and with IA strategy.

about 10% in weight of the total amount of attached-growth biomass as estimated by Lim et al. [23] in an earlier study involving the use of MBSBR with 8% (v/v) of 8-mL PU foam cubes for nitrogen removal, it was vital for the protection of basal layers and limited the degradation rate of inner biomass [39]. Zhou et al. [40] suggested that the biomass presented in the deeper layer became even more active although the microbial activities were inhibited on the outer layer; thus, justifying higher nitrification rate in Phase IV. In comparison, the PE cylindrical rings as the support media possessed only the outer layer for the growth of biomass which failed to benefit the operation of MBSBR once this outer biomass was inhibited by the toxic pollutants in wastewater.

3.3. Phases V, VI, and VII studies: Comparison of the performance of MBSBRs with IA and IASF strategies

The concentration profiles of the nitrogen species for reactors R-IA and R-IASF in Phases V and VI are shown in Figs. 4 and 5, respectively. The TN removal efficiencies were calculated from the profile studies in the respective Phases and shown in Table 5. Fig. 4 shows that complete removal of NH_4^+-N was achieved for both MBSBRs in Phase V. The advantage of incorporating SF mode during the REACT period is seen in the TN removal of almost 80% in R-IASF against 64% in R-IA in Phase V. In general, the SF mode will permit better exploitation of carbon source in the influent to stimulate SND process. The introduction of influent during the non-aeration periods of R-IASF would diminish the potential of carbon source originated from domestic wastewater from being oxidized in the mixed liquor. Indeed, this carbon source was primarily used to reduce the oxidized nitrogen. In contrast, a single feeding mode adopted for R-IA would cause an unnecessary waste of carbon source from domestic wastewater as this reducing agent would be thoroughly oxidized mainly amid the first aeration period by heterotrophic bacteria. The subsequent removal of TN in R-IA was probably attributed to the utilization of carbon source by the denitrifying bacteria from the endogenous respiration as explained by Wang et al. [41].

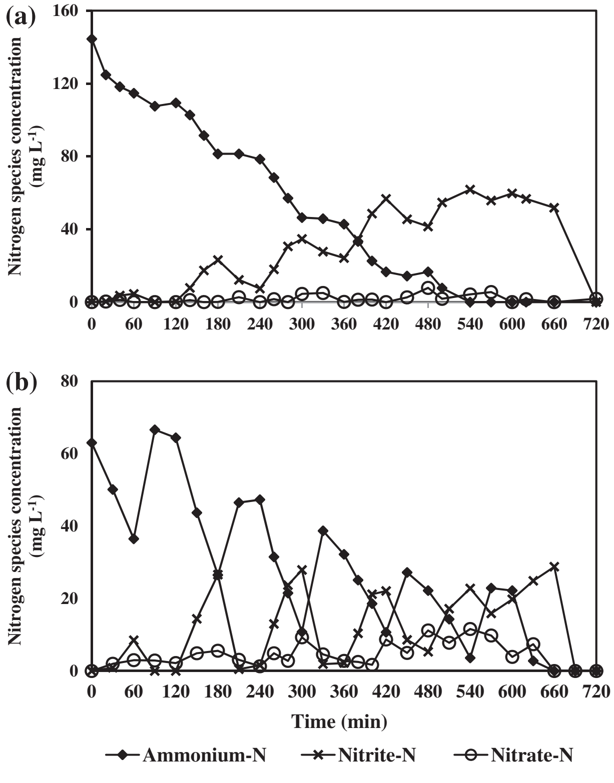

Fig. 4. Concentration profiles of nitrogen species during the REACT period for reactors (a) R-IA and (b) R-IASF operated at leachate volumetric ratio of 14.3% and with IA strategy.

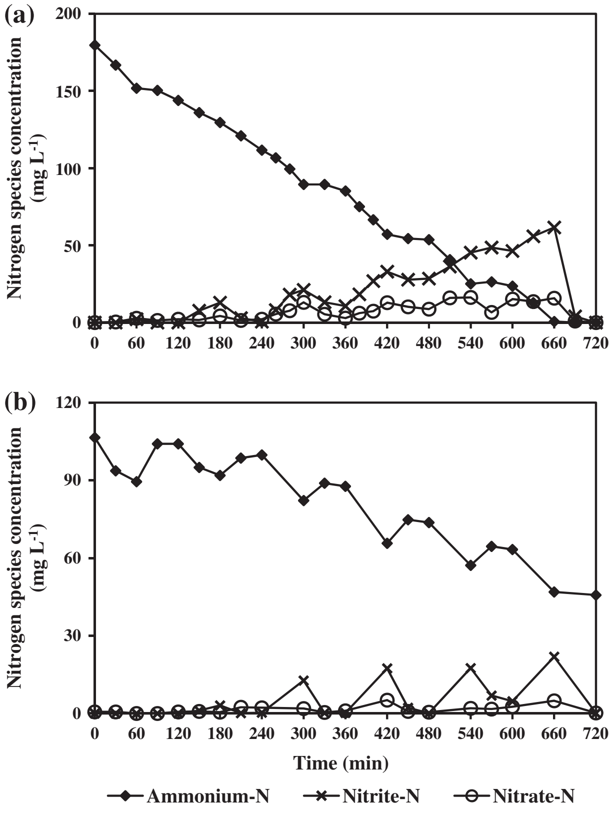

Fig. 5. Concentration profiles of nitrogen species during the REACT period for reactors (a) R-IA and (b) R-IASF operated at leachate volumetric ratio of 20.0% and with IA strategy.

Table 5
Concentrations of nitrogen species in mixed liquor and TN removal efficiencies during the REACT period prior to the addition of ethanol solution

Reactor	Phase	Concentration (mg L^{-1})			TN removal (%)
		NH_4^+-N	NO_2^--N	NO_3^--N	
R-IA	V	0	51.8	0	64.2
	VI	0	61.6	15.8	69.7
R-IASF	V	0	28.8	0	79.8
	VI	45.7	21.9	4.9	71.0

The increase in leachate volumetric ratio to 20% in Phase VI had resulted in the poor performance of reactor R-IASF as evidenced by the incomplete removal of NH_4^+-N (Fig. 5(b)). In addition, the TN removal of R-IASF in Phase VI deteriorated to around 71%, which was the same as that of R-IA (Table 5). The poor performance of R-IASF could be explained by the fact that the biomass was acclimated to a fairly low concentration of NH_4^+-N in the previous Phase (about 60 mg L^{-1} as depicted in Fig. 4(b)) due to SF mode and was therefore inhibited when the NH_4^+-N concentration was almost doubled in Phase VI (Fig. 5(b)). In contrast, Fig. 5(a) shows that complete NH_4^+-N removal was still achievable before reaching the end of REACT period in R-IA in Phase VI. The greater tolerance of R-IA to higher NH_4^+-N concentration was expected because of better acclimation to higher NH_4^+-N concentration in the previous Phase.

When the volumetric ratio of leachate in influent was further increased to 25% in Phase VII, reactor R-IA failed as incomplete removal of NH_4^+-N was observed in the effluent (data not included). Thus, the volumetric ratio of leachate in the combined stabilized landfill leachate and domestic wastewater is the dominant factor in the unsustainability of R-IA operation.

4. Conclusions

At the leachate volumetric ratio of 14%, the performance of the two MBSBRs packed with PU and PE, respectively, was comparable with complete NH_4^+-N removal, but the SBR failed to remove NH_4^+-N completely under the CA strategy. When the aeration strategy was changed from CA to IA, the performance of MBSBR packed with PE rings deteriorated with incomplete NH_4^+-N removal. In contrast, the IA strategy was beneficial for MBSBR packed with PU cubes with the enhancement of the rate of NH_4^+-N removal. For the MBSBR with PU media, a TN removal efficiency of 80% was attained by coupling the IA strategy with SF mode compared to an efficiency of 64% in the MBSBR with only IA strategy. Further increase in leachate volumetric ratio to 20% had brought about incomplete NH_4^+-N removal in the MBSBR with IA-SF operation in comparison to the MBSBR operated with only IA strategy which could still achieve complete removal of NH_4^+-N.

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