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# Advanced nitrogen removal *via* nitrite from municipal landfill leachate using a two-stage UASB–A/O system<sup>\*</sup>





Lina Wu<sup>1,2</sup>, Yongzhen Peng<sup>3,\*</sup>, Xiao Shi<sup>1</sup>, Chengyao Peng<sup>3</sup>, Jie Zhang<sup>2</sup>

<sup>1</sup> Beijing Institute of Petrochemical Technology, Beijing 102617, China

<sup>2</sup> School of Municipal and Environmental Engineering, Harbin Institute of Technology, Harbin 150090, China

<sup>3</sup> Key Laboratory of Beijing for Water Quality Science and Water Environment Recovery Engineering, Beijing University of Technology, Beijing 100124, China

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# ABSTRACT

A system consisting of a two-stage up-flow anaerobic sludge blanket (UASB) reactor and an anoxic/aerobic (A/O) reactor was used to treat municipal landfill leachate. Denitrification took place in the first stage of the UASB reactor (UASB1). The chemical oxygen demand of the UASB1 effluent was further decreased in the second stage (UASB2). Nitrification was accomplished in the A/O reactor. When diluted with tap water at a ratio of 1:1, the ammonia nitrogen concentration of the influent leachate was approximately 1200 mg·L<sup>-1</sup>, whereas that of the system effluent was approximately 8–11 mg·L<sup>-1</sup>, and the corresponding removal efficiency is about 99.08%. Stable partial nitrification was achieved in the A/O reactor with 88.61%–91.58% of the nitrite accumulation ratio, even at comparatively low temperature (16 °C). The results demonstrate that free ammonia (FA) concentrations within the 1–30 mg·L<sup>-1</sup> range, partial nitrification could be achieved, whereas when FA exceeded 280 mg·L<sup>-1</sup>, the nitrification process was entirely inhibited. Temperature was not the key factor leading to partial nitrification within the 16–29 °C range. The inhibitory influence of free nitrous acid (FNA) on nitrification was also minimal when pH was greater than 8.5. Thus, FA concentration was a major factor in achieving partial nitrification.

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# 1. Introduction

Leachate is defined as wastewater with a high concentration of organic matter [1]. The composition of landfill leachate is very complicated, and its attributes and quantity can vary considerably depending on the time elapsed since waste disposal in the landfill [2]. The main characteristics of landfill leachate are high concentrations of ammonia nitrogen (NH<sub>4</sub><sup>+</sup>-N) and organic matter, which cause great difficulties in treating leachate [3]. Some research has indicated that a high ammonia nitrogen content can inhibit bacterial activity [4]. Therefore, some physicochemical processes are used to decrease the concentration of ammonia nitrogen to a level that is suitable for subsequent biological processes [5]. For instance, ammonia stripping, powdered activated carbon-sequencing batch reactor (SBR) and coagulation–sedimentation processes effectively removed organic compounds and nitrogen from

Corresponding author.

leachate [6]. However, such physicochemical processes have many drawbacks, such as high operational costs and high agent consumption.

Biological denitrogenation can meet the important need to remove nitrogen contamination from landfill leachate while avoiding the above problems. One of the most effective measures for biological denitrogenation is to achieve stable partial nitrification [7]. Compared with traditional processes for biological nitrogen removal, partial nitrification can reduce the consumption of oxygen by 25% for nitrification and carbon source by 40% for denitrification [8]. In the majority of the published papers about the conventional nitrogen removal technologies, an ammonia nitrogen removal efficiency of greater than 90% could not be achieved [9]. Especially, the total nitrogen (TN) removal efficiency is lower. Nitrate denitrification requires a  $\rho(COD_{Cr})/\rho(TKN)$ ratio greater than 4, whereas nitrite denitrification can be achieved with a ratio of 2.5 or more. Thus, partial nitrification requires a less concentrated carbon source than traditional nitrification. The key to achieving partial nitrification is inhibiting the activity of nitrite-oxidizing bacteria (NOB) such as Nitrobacter so that ammonia-oxidizing bacteria (AOB) such as Nitrosomonas dominate the nitrifying bacterial community [10]. High temperature is a commonly required condition to inhibit the activity of NOB and thus achieve stable partial nitrification [11-13]. Yoon and Kim [14] reported that FA could inhibit both AOB and NOB, but to different extents. NOB are more sensitive to FA than

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E-mail addresses: wlncyj@sina.com, pyz@bjut.edu.cn (Y. Peng).

are AOB: FA concentrations of 0.1–1.0 mg $\cdot$ L<sup>-1</sup> inhibit NOB, whereas only much higher concentrations of  $10-150 \text{ mg} \cdot \text{L}^{-1}$  inhibit AOB [15–18]. Therefore, when FA concentration exceeds the threshold for NOB inhibition, but remains within the AOB tolerance range, nitrite will accumulate in the reactor [19]. Once the FA concentration exceeds the AOB tolerance level, it inhibits both NOB and AOB, causing nitrification to cease. Therefore, it is very important to maintain FA concentration within the range that inhibits NOB but not AOB so that partial nitrification can be achieved and maintained. Similarly, to other bacterial groups, temperature also affects the activities of the nitrifying bacterial community. Nitrification is promoted at higher reactor temperature. At temperatures above 25 °C, the growth rate of AOB is higher than that of NOB [20], such that partial nitrification is easily achieved and maintained. Therefore, temperature is considered a factor in nitrite accumulation [21]. However, few studies have reported that partial nitrification was successfully achieved and maintained at low temperatures (16–18 °C) [22]. FNA concentration is reported to be an additional factor affecting partial nitrification [23]. Vadivelu et al. [24] reported that synthetic metabolism of NOB was inhibited at FNA concentration  $>0.011 \text{ mg} \cdot \text{L}^{-1}$ , and that the synthetic metabolism of microorganisms was completely inhibited when FNA reached 0.023  $mg \cdot L^{-1}$ . Hence, NOB activity was selectively inhibited by FNA concentrations between  $0.011 \text{ mg} \cdot \text{L}^{-1}$  and  $0.1 \text{ mg} \cdot \text{L}^{-1}$ .

Although many studies have examined the factors affecting the achievement of partial nitrification, most of them used synthetic rather than municipal wastewater. In addition, few previous studies reported complete nitrogen removal *via* nitrite with nitrite accumulation greater than 88%, especially when the ammonia nitrogen concentration  $(NH_4^+-N)$  of the system influent was higher than 1200 mg·L<sup>-1</sup>. Furthermore, few studies have achieved stable and effective partial nitrification at low temperatures (16–18 °C) using an anaerobic/aerobic system to treat municipal wastewater [25].

Based on the above research background, the biological treatment system, consisting of a two-stage UASB reactor and an anoxic/aerobic (A/O) reactor, was a fully relied upon biological process to treat the municipal landfill leachate without any physicochemical pretreatment. This experiment also investigated the effects of FA, temperature, and FNA on partial nitrification through a treatment of municipal landfill leachate containing high concentrations of ammonia nitrogen specifically at low temperature. Other factors related to partial nitrification were also explored.

# 2. Materials and Methods

Municipal landfill leachate discharged from the Liulitun landfill site in Beijing, China was collected for the study. Table 1 summarizes the characteristics of the leachate.

A system comprising a two-stage up-flow anaerobic sludge blanket (UASB) reactor and an anoxic/aerobic (A/O) reactor was used to treat landfill leachate. Part of the recycled effluent as well as the influent of the system was pumped into the first-stage UASB reactor (UASB1). The organic compounds in the raw landfill leachate were depleted by serving as the carbon source for denitrification of the recycled effluent in UASB1, while simultaneously, methanogenesis occurred. Some organic compounds in the UASB1 effluent were subsequently depleted *via* methanogenesis in UASB2. Recycled sludge from the clarifying tank was then pumped into the anoxic zone (the first chamber of the A/O reactor) in which denitrification of NO<sub>x</sub>–N (nitrite and nitrate nitrogen) took place. Nitrification of ammonia occurred in the aerobic zone

of the A/O reactor. A diagram of the process is presented in Fig. 1. The effective volume of UASB1 was 4.25 L, and that of UASB2 was 8.25 L. The A/O reactor had a working volume of 15 L, which was divided into ten chambers, with the first chamber forming the anoxic zone and the remaining nine the aerobic zone.

The experiment was conducted in three phases over 90 days. In Phase I, partial nitrification was not achieved at high temperature (30 days); in Phase II, partial nitrification was achieved at high temperature (30 days); and in Phase III, partial nitrification was achieved at low temperature (30 days). The inhibitory effect of excessively high FA level on partial nitrification as well as on all nitrification was investigated in Phase I, and the favorable FA concentration range within which partial nitrification was obtained was investigated in Phases II and III. FNA was also investigated in these phases.

The operational conditions were as follows: the influent flow rate of the two-stage UASB-A/O system was 3 L·day<sup>-1</sup>. The effluent recycling ratio was 300%, and the sludge recycling ratio was 100%. The operational temperatures of UASB1 and UASB2 were maintained at 30 and 35 °C, respectively, by a heater and thermostat. The temperature of the A/O reactor was maintained at 27-29 °C by a heater during Phases I and II. In Phase III, the heater was removed, and the A/O reactor was operated at ambient temperature (16–18 °C). The dissolved oxygen (DO) level in the second to tenth chambers of the A/O reactor varied from 0.5 to 5 mg  $\cdot$  L<sup>-1</sup>. pH ranged from 8 to 9 in the ten A/O reactor chambers. Sludge retention time (SRT) was maintained at approximately 30-40 days. The mixed liquor suspended solid (MLSS) concentration remained within the range 3000–5000 mg $\cdot$ L<sup>-1</sup>. No supplementary source of carbon was added to the system. The sampling points were as follows: raw leachate (raw), UASB1 influent (mix) and effluent (U1e), UASB2 (U2e) effluent, the anoxic zone of the A/O reactor (A1), and the remaining nine individual chambers of the aerobic zone of the A/O reactor (02-010).

COD,  $NH_4^+$ -N,  $NO_3^-$ -N and  $NO_2^-$ -N were measured according to standard methods [26]. TN was analyzed by a multi N/C 3000 TOC analyzer (Analytik Jena AG, Germany). DO and pH were monitored using a dissolved oxygen meter (WTW DO 330i, Germany) and a pH meter (WTW pH 340i, Germany).

# 3. Results and Discussion

#### 3.1. Phase I: Partial nitrification not achieved at high temperature

The ammonium nitrogen concentration of the raw leachate was approximately 2500 mg·L<sup>-1</sup>, a level that may inhibit bacterial activity, especially at high pH [27]. Fig. 2 illustrates the typical variation in nitrogen concentrations in the system during the first phase. When the influent ammonium nitrogen concentration was  $2500 \text{ mg} \cdot \text{L}^{-1}$ , the concentration in the A/O reactor influent was  $928 \text{ mg} \cdot \text{L}^{-1}$ , due in part to dilution by the recycled effluent and sludge. The ammonium nitrogen concentration of the final effluent was close to  $800 \text{ mg} \cdot \text{L}^{-1}$ . This decrease can be attributed to high pH and alkalinity, which lead to ammonium stripping, as well as the removal of ammonium by simultaneous nitrification and denitrification (SND). In this phase, the temperature in the A/O reactor was 27-29 °C, which is the optimal temperature range for partial nitrification. However, partial nitrification was not achieved, which was attributed to the inhibition of nitrifying bacteria by FA.

Fig. 2 shows the typical variation in nitrogen in the system during this phase. The COD concentration of the influent was 11000 mg  $\cdot$  L<sup>-1</sup> and COD removal efficiency was 85.45%.

Table 1
Characteristics of raw leachate

Parameter	Chemical oxygen demand $(COD)/mg \cdot L^{-1}$	Ammonia nitrogen (NH₄+N)/mg·L <sup>-1</sup>	Total phosphate (TP)/mg⋅L <sup>-1</sup>	Total nitrogen (TN)/mg·L <sup>-1</sup>	Nitrite and nitrate nitrogen $(NO_x^{-}-N)/mg \cdot L^{-1}$	рН
Range	8000-11000	2200-2500	9–15	2230-2530	0.5–15	7.2–7.9



Fig. 1. Flow diagram of the two-stage UASB-A/O system.



Fig. 2. COD and nitrogen variation during Phase I. Sampling point: 1-raw; 2-mix; 3-U1e; 4-U2e; 5-A1; 6-O2; 7-O3; 8-O5; 9-O7; and 10-O10.

# 3.2. Phase II: Partial nitrification achieved at high temperature

For Phase II, the leachate used in the first phase was diluted with tap water at a ratio of 1:1. Fig. 3 shows the typical variation in nitrogen in the system during Phase II.

In this phase, the influent ammonia nitrogen concentration was 1200 mg·L<sup>-1</sup>, due to dilution with recycled effluent, the concentration in the UASB1 effluent was 311 mg·L<sup>-1</sup>. The ammonia nitrogen concentration of the A/O reactor influent was less than 150 mg·L<sup>-1</sup>, due to the further dilution of the recycled sludge. Ammonia nitrogen was removed by nitrification in the second chamber of the A/O reactor, which served as the first chamber of the aerobic zone. Nitrification ceased in the fifth chamber. Partial nitrification was almost completed. The final ammonia nitrogen concentration of the A/O reactor effluent was 8 mg·L<sup>-1</sup>, corresponding to an ammonia nitrogen removal efficiency of 99.33%. The NO<sub>3</sub><sup>-</sup>-N concentration of the final effluent was 16 mg·L<sup>-1</sup> and that of NO<sub>2</sub><sup>-</sup>-N was 174 mg·L<sup>-1</sup>. Stable partial nitrification was thus achieved and maintained with 91.58% nitrite accumulation in this phase.

The COD concentration of the influent was within the range 4400 mg·L<sup>-1</sup>-4600 mg·L<sup>-1</sup>, and the average COD/TN ratio was less than 4. Traditional denitrification methods require a COD/TN ratio of



Fig. 3. Nitrogen variation during Phase II. Sampling point: 1-raw; 2-mix; 3-U1e; 4-U2e; 5-A1; 6-O2; 7-O3; 8-O5; 9-O7; and 10-O10.

more than 4, whereas partial nitrification–denitrification can be achieved at a ratio of 2.5 or more [28]. Although the original leachate had poor biodegradability, the ammonia nitrogen was removed by partial nitrification, and the carbon source was sufficient, requiring no supplementation. The effluent COD of the UASB1 was less than 1700 mg·L<sup>-1</sup>. The organic matter within the landfill leachate served as the carbon source for denitritation of the recycled effluent in UASB1. Supplementary carbon was not required, as the carbon content of the influent was sufficient. However, other denitrification processes require the addition of carbon to the leachate [29]. Most of the organic matter was removed by denitrification and methanogenesis in the two-stage UASB. The low biodegradable COD benefited the complete nitrification in the A/O reactor. The final effluent COD of the system of this phase was approximately 1300 mg·L<sup>-1</sup>.

### 3.3. Phase III: Partial nitrification achieved at low temperature

The characteristics of the influent used in Phase III were the same as for the second phase. The heater was removed from the A/O reactor in this phase. Because the experiment was conducted in winter, the temperature in the A/O reactor was decreased to 16-18 °C. Fig. 4 shows the typical variation of nitrogen in the system during this phase. Ammonia nitrogen was not depleted in UASB1 or UASB2. Due to dilution by recycled sludge, the ammonia nitrogen concentration of the A/O reactor influent was decreased to 110-130 mg·L<sup>-1</sup>. The NO<sub>2</sub><sup>-</sup>-N and NO<sub>3</sub><sup>-</sup>-N in the recycled sludge were removed by denitrification in the anoxic zone of the A/O reactor. The ammonia nitrogen concentration of the system influent was 1200 mg $\cdot$ L<sup>-1</sup>, which was the same as in the second phase. The ammonia nitrogen concentration of the system effluent was 11 mg $\cdot$ L<sup>-1</sup>, giving 99.08% removal efficiency of ammonia nitrogen. In the final effluent, NO<sub>3</sub><sup>-</sup>-N concentration was  $18-28 \text{ mg} \cdot \text{L}^{-1}$ , and that of  $NO_2^-$ -N was 140–220 mg·L<sup>-1</sup>. Nitrite accumulation in Phase III was lower than in Phase II but still as high as 88.61%-88.72%. Although the temperature in the A/O reactor was 10 °C lower than in Phases I and II. stable partial nitrification was still achieved in this phase.

Many studies have reported that ammonia nitrogen removal efficiency decreases sharply below 20 °C [30,31]. During Phase III, the temperature in the A/O reactor was decreased to 16–18 °C, nevertheless, efficient and stable (99% removal efficiency) elimination of ammonia nitrogen was achieved. Compared with other forms of leachate treatment, extremely high removal efficiency of ammonia nitrogen (99.08%), was obtained by using the biological system, without any form of pretreatment or physicochemical treatments. The results suggest that this combined process provides an economic means of removing ammonia nitrogen from leachate.

Many previous studies reported that partial nitrification was not achieved at temperatures less than 20 °C, because NOB have higher activity than AOB below 25 °C. However, in the present study, nitrite accumulation in Phase III was still as high as 88.61% when the temperature dropped to 16–18 °C. The results clearly demonstrate that the unfavorable effects of low temperature on achieving and maintaining nitritation, especially partial nitrification, were successfully overcome *via* the proposed method.

The COD degradation of this phase was similar to that of Phase II. The influent COD concentration was approximately 4400–4600 mg·L<sup>-1</sup>, and that of the final effluent was kept within the range 1200–1300 mg·L<sup>-1</sup>. This residual COD was mostly refractory organic matter and could not be removed by microbial treatment.

# 3.4. Comparison of the three phases

As shown in Table 2, the operational conditions of the three phases were similar except that the influent ammonia nitrogen concentration was higher for the first phase than for the other two and the temperature of the third phase was 10 °C lower than in Phases I and II. Despite the lower temperature of Phase III, stable partial nitrification was still obtained, with 88.61% nitrite accumulation and 99.08% ammonia nitrogen removal efficiency. It is therefore concluded that temperature was not the key factor at 16–29 °C in achieving stable partial nitrification and high ammonia nitrogen removal efficiency.

The effects of FA and FNA on partial nitrification were studied to further investigate the factors leading to partial nitrification.

The concentrations of FA and FNA were calculated as shown in Eqs. (1) and (2) [32], where  $\rho$  represents concentration (mg·L<sup>-1</sup>) and *T* is temperature (°C).

$$\rho_{(\text{FA})} = \frac{\rho_{(\text{NH}_{4}^{+}-\text{N})} \times 10^{\text{pH}}}{\exp[6334/(273+T)] + 10^{\text{pH}}}$$
(1)

$$\rho_{(\text{FNA})} = \frac{\rho_{(\text{NO}_{2}^{-}\text{N})}}{\exp[-2300/(273+T)] \times 10^{\text{pH}}}$$
(2)

The concentrations of FA and FNA were the two key factors in achieving and maintaining partial nitrification.

The variations in FA and FNA are presented in Fig. 5. In the first phase, the FA concentration in each chamber exceeded 280 mg·L<sup>-1</sup>.



Fig. 4. Nitrogen variation during Phase III. Sampling point: 1-raw; 2-mix; 3-U1e; 4-U2e; 5-A1; 6-O2; 7-O3; 8-O5; 9-O7; and 10-O10.

Table 2
Operational conditions and results of the A/O reactor during the three phases

Phase	Flow rate/L·d <sup>-1</sup>	T/°C	Effluent recycling ratio/%	Influent NH <sub>4</sub> <sup>+</sup> -N/mg·L <sup>-1</sup>	$NH_4^+$ -N loading rate (ALR)/kg·m <sup>-3</sup> ·d <sup>-1</sup>	Hydraulic detention time (HRT)/d	Return sludge ratio/%	Nitrite accumulation/%	Effluent NH <sup>+</sup> <sub>4</sub> -N/mg·L <sup>-1</sup>
Phase I	11	27-29	300	928	0.8	1.4	100	0	800
Phase II	11	27-29	300	110	0.08	1.4	100	93	8
Phase III	11	16-18	300	110	0.08	1.4	100	88	11

Nitrification did not occur during this phase, despite the optimal temperature of 27–29 °C. Thus, excessive FA concentration caused by high ammonia nitrogen not only partially inhibited nitrification but also caused it to cease completely. In this phase, FNA concentrations were less than 0.011 mg·L<sup>-1</sup>. Hence, the activity of NOB was not inhibited by FNA.

In Phase II, the ammonia nitrogen concentration of the influent was decreased by dilution with tap water, thereby lowering FA concentration (1.74–28.4 mg·L<sup>-1</sup>; see Fig. 5). pH remained above 8.5, and the FA concentration was still 1 mg·L<sup>-1</sup> in the tenth chamber of the A/O reactor. Because of this, the FNA concentration was below 0.011 mg·L<sup>-1</sup>. The effect of FNA on NOB can therefore be neglected. In the second phase, the FA concentration inhibited NOB but not AOB. Thus, stable partial nitrification was obtained.

During the third phase, the FA concentration in the first chamber of the A/O reactor was still 18.3 mg·L<sup>-1</sup>, as shown in Fig. 5, although the temperature was 10 °C lower than that of the second phase. The FA concentration gradually declined as a result of the decreasing ammonia nitrogen concentration and pH along with nitrification, but the lowest FA concentration of 1.48 mg·L<sup>-1</sup> still effectively inhibited NOB. Hence, stable partial nitrification was accomplished with 88.61% nitrite accumulation despite the lower operating temperature. The original FA concentration dropped slightly in the A/O reactor under the lower temperature. Stable partial nitrification was obtained in this phase

because the ammonia nitrogen concentration was reduced by the dilution of the raw leachate. Furthermore, the reduction of the FA concentration prevented the inhibition of nitrification, that was observed when the FA level was excessively high; as a result, nitrification was completed. The COD of the raw leachate was high, indicating that it was rich in organic matter. The nitrite and nitrate in the recycled effluent from the A/O reactor were entirely denitrified in UASB1. Furthermore, the original alkalinity concentration in the A/O reactor was 2000 mg CaCO<sub>3</sub> $\cdot$ L<sup>-1</sup>. This alkalinity, which was adequately supplied by completed denitrification in the A/O reactor, was enriched. The "ammonia valley" (the local minimum in the pH profile) still maintained a pH higher than 8.5; as a result, the FA concentration decreased no faster than the temperature. FA concentrations were always greater than  $1 \text{ mg} \cdot \text{L}^{-1}$  in all chambers of the A/O reactor. Thus, FA inhibited NOB in all chambers of the A/O reactor, and stable partial nitrification was obtained under the lower temperature. The pH value in Phase III was the same as that in the second phase. Therefore, the FNA concentration was so low that the effect of FNA on the activity of NOB could be neglected.

Based on the above analysis, stable and effective partial nitrification took place in the A/O reactor during Phases II and III because the FA concentration was adjusted to a favorable range *via* influent dilution. Within this range, the FA concentration inhibited NOB but not AOB, and thus NOB were gradually washed out in the A/O reactor, optimizing



Fig. 5. FA and FNA variations in the A/O reactor during different phases.

the composition of the nitrifying bacterial community. Hence, it was concluded that FA was the primary factor in achieving and maintaining stable partial nitrification, and that temperature was a secondary factor. The effect of FNA on partial nitrification could be neglected at pH >8.5.

#### 4. Conclusions

The following conclusions are drawn from the experimental results:

- (1) The two-stage UASB–A/O process was efficient for landfill leachate treatment. The ammonia nitrogen concentration of the influent could be reduced from 1200 mg·L<sup>-1</sup> to 8–11 mg·L<sup>-1</sup>, and a removal efficiency of 99.08% was achieved even at low temperature (16–18 °C). Specifically, stable partial nitrification was achieved with more than 88.61% nitrite accumulation at low temperature (16 °C). There are few reports of similar results using other biological treatments of leachate.
- (2) In Phase I, FA concentration exceeded 280 mg·L<sup>-1</sup>, which can inhibit both NOB and AOB. Consequently, partial nitrification could not take place, despite the favorable temperature range of 27–29 °C in the A/O reactor. In Phases II and III, FA concentration in the A/O reactor decreased (range 1.48–28.4 mg·L<sup>-1</sup>) due to reduction of the influent ammonium nitrogen concentration by diluting raw leachate 1:1 with tap water. FA inhibited only NOB but not AOB. As a result, stable partial nitrification was obtained with 88.61%–91.58% nitrite accumulation in the A/O reactor and 99.08% nitrogen removal efficiency in the whole system, despite the comparatively low temperature (16–18 °C).
- (3) The three phases of the experiment demonstrate that FA concentration was the major factor in achieving stable partial nitrification of leachate. Hence, it is possible to control the FA concentration to achieve and maintain stable partial nitrification in practical engineering for landfill leachate treatment, even at low temperature (16–18 °C).

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