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Leachate treatment using a demonstration aged refuse biofilter

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Abstract

Approximately 7000 m^3 of aged refuse (AR) with a placement of over eight years was excavated from Shanghai Refuse Landfill, the largest landfill in China, and used for the construction of a two-stage bioreactor (AR biofilter) media for the biological treatment of 100 m^3 of refuse landfill leachate. It was found that over 64% of COD, 96.9%–99.8% of NH_4^+-N, and 95.8%–99.8% of BOD_5 could be removed by the AR biofilter, when the leachate with initial COD, BOD_5, and NH_4^+-N concentrations were 986–4128 mg/L, 264–959 mg/L, and 538–1583 mg/L, respectively. The corresponding concentrations in the effluent were reduced to below 300–400 mg/L, 2–12 mg/L, and 10–20 mg/L, respectively. The effluent was clear and pale yellow with suspended solid below 150 mg/L and color below 150 Pt/Co degree. Meanwhile, the total nitrogen removal was only 49%–63%, indicating a relative poor denitrification capacity of AR biofilter. The effluent pH was neutral and the population of *Escherichia coli* was less than 10^{-1} CFU/mL. Hence, it was considered that the demonstration project can work well for the effective treatment of leachate.

Key words: aged refuse; landfill; leachate treatment; COD; ammonia

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Introduction

Municipal landfilling is still the most widely applied method for solid waste disposal in the world. A significant consequence of the present waste disposal practice is the generation of landfill leachate, which brings about special environmental concern. Landfill leachate is liquid that percolates through waste and extracts dissolved or suspended materials. A significant quantity of chemical constituents enters into leachate when water passes through solid wastes which are undergoing decomposition in the landfill compartment (George et al., 1993). It is a complex wastewater containing a great amount of refractory organic matters, which are difficult to be degraded biologically by conventional methods. Consequently, leachate treatment is one of the most important tasks in refuse landfill management (Zhao, 1999).

At present, technologies for the leachate treatment include biological treatment, physical-chemical process, bioreactor technique and natural systems such as constructed wetlands. In view of the feasibility, effectiveness and economy, biological methods which consist of activated sludge process (Bae et al., 1999), membrane bioreactor process (Bodzek et al., 2006), upflow anaerobic sludge blanket (UASB) process (Kennedy and Lentz, 2000), and sequencing batch reactor (SBR) process (Klimiuk and Kulikowska, 2006) etc. play important roles for leachate treatment. Due to the specialty of leachate, biological treatment may perform a relatively reasonable treatment effect (Castillo et al., 2007), but it can hardly achieve high and reliable ammonia and COD removal efficiencies alone, especially for the stabilized leachate (Canziani et al., 2006). Physical-chemical techniques (flocculation/precipitation; adsorption; evaporation; membrane technologies and chemical oxidation etc.) are simple in process, easy to operate, and insensitive to temperature change. However, the high operational cost limits their universal application (Kurniawan et al., 2006). In most cases, physical-chemical methods are used as pre-treatment to improve the biodegradability or post-treatment to meet stricter discharge standard (Kurniawan et al., 2006; Tatsi et al., 2003). Bioreactor technique (Reinhart et al., 2002) controls leachate pollution by recirculating leachate *in situ* for the degradation of biodegradable organic matters. However, NH_4^+-N is a significant long-term pollution problem in landfills (Kjeldsen et al., 2002). Laboratory results demonstrate that *in situ* NH_4^+-N removal is feasible in bioreactor landfills, but more field-scale verifications are needed (Berge et al., 2006, 2007a, 2007b). Constructed wetlands are a low-cost alternative for leachate treatment, resulting in a treated effluent suitable for irrigation in agriculture or discharge to nearby environment (Sawaittayothin and Polprasert, 2007). However, constructed wetlands may not be a competitive option compared to other treatment methods where space is a major constraint.

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An innovative process for leachate treatment using a biofilter consisting of 8–10 year old aged refuse was initiated (Zhao et al., 2002). The laboratory results show that initial chemical oxygen demand (COD), biochemical oxygen demand (BOD) and NH_4^+-N reduced from 3000–7000, 540–1500, and 500–800 mg/L to lower than 100–350, 10–200, and 10–25 mg/L, respectively. Such a biofilter is also very effective for the treatment of sewage (Zhao et al., 2007) and feedlot wastewater (Zhao and Shao, 2004).

Based on our previous work and the construction of low rated trickling filter, a demonstration engineering project of this simple and high cost-effective leachate treatment process, which can manage 100 m^3 leachate per day, was conducted in Shanghai Refuse Landfill. The objective of this article was to investigate the long-term leachate treatment behavior of scaling-up aged refuse (AR) biofilter through the evaluation of the removal of color, suspended solid (SS), organic pollutants (COD, BOD_5, DO, etc.), and nitrogen (NH_4^+-N, NO_3^--N, NO_2^--N, TN) by the AR biofilter. This research may serve as guidance for other demonstration projects on the AR biofilter, providing a more informed approach to designing and operating the AR biofilter in landfills in the future.

1 Materials and methods

1.1 Biofilter materials – aged refuse

The compositions of the refuse in Shanghai Refuse Landfill were determined to be 40% moisture, 12% plastic films, bags, bottles, and other products, 24% organic matter such as trees, woods, and cooking wastes, and 24% inorganic matter such as stone, sand, coal ashes, and glass bottles (Zhao et al., 2002). The aged refuse (AR) used as biofilter materials was excavated from the same landfill compartment which had been closed for eight years, as described in our published article (Zhao et al., 2002). After eight years of placement and stabilization, the weight percentage of refuse with different diameters, > 40 mm, 15–40 mm, and < 15 mm, accounted for 29.50%, 32.85%, and 37.65%, respectively. Considering the operability of engineering, refuse was screened with limiting diameter less than 40 mm instead of 15 mm used in the laboratory and introduced as two-stage biofilter materials. Around 70.5% weight of the total excavated refuse can be used for the feed materials of the biofilter, in which the larger sizes of nondegradable matters such as larger stones, glass bottles, plastic films (bags), rubbers etc., were absent.

1.2 Two-stage AR biofilter

As can be seen from Fig. 1, the AR biofilter was separated into two stages. The cross section of each stage was trapezoid. The length of two parallel sides and vertical height of the trapezoid were 59, 55 and 3 m, respectively. The width of each biofilter was 45 m with about 7000 m^3 screened AR contained inside.

Landfill leachate was pumped from leachate pond and sprayed over the first-stage biofilter. After passing through

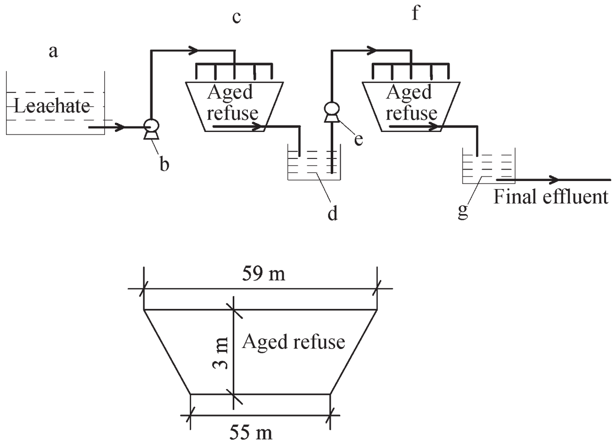

Fig. 1 Two-stage AR biofilter. (a) leachate pond; (b) first pump; (c) first-stage biofilter; (d) first collection basin; (e) second pump; (f) second-stage biofilter; (g) second collecting basin.

the first-stage biofilter due to gravity, leachate was collected in the first collection basin and then pumped and sprayed over the second-stage biofilter. The final effluent was collected in the second collecting basin. A rotary distributor system was adopted for discharging the leachate onto the surface of biofilter. In each biofilter, the rotary distributor consisted of eight hollow horizontal center columns, each of which carried 11 arms containing a number of nozzles. All of these nozzles point in the same direction at right angles to the arms and the reaction of the discharge through them caused the arms to revolve. The leachate was pumped by the first pump and second pump and sprayed over the surface of two stages with the same frequency, 10 times per day and each time for 30 min. The flow quantity of influent leachate was 100 m^3 per day.

1.3 Sampling and analyzing of AR and leachate

For chemical analysis, AR in the two-stage biofilter was sampled and air-dried at room temperature, and then grinded into powder and sieved through a 0.25 mm-pore screen. The pH of the AR sample was measured in 0.01 mol/L $CaCl_2$ solution (refuse:solution, 1:2.5, *m/m*) with pH meter (Leici Instrument Co., Shanghai, China). Content of organic matters was determined by oxidation with potassium dichromate in a strong acid open medium and back titration by ferrous sulphate in the presence of an indicator (Pansu and Gautheyrou, 2006). Aerobic bacteria and anaerobic bacteria which can approximately reflect the activity of different microorganisms inside the AR biofilter were analyzed by standard methods (APHA, 1998).

Leachate from the collecting basin was analyzed periodically during the 191 days period since 1st December 2006. Sampling of leachate was carried out not in rainy days to avoid dilution by the rain. Leachate samples were taken from the leachate collecting basin three times a day with an interval of at least two hours and mixed together as one sample to ensure the representativeness of each sample. Concentrations of COD, NH_4^+-N, DO and pH value were monitored every five days and concentrations of NO_3^--N, NO_2^--N, BOD_5, and SS were analyzed every 23 days. All these data were analyzed according to standard methods (APHA, 1998) and in duplicate.

2 Results and discussion

2.1 Characterization of AR

Table 1 shows the basic parameters of AR used as biofilter materials. It can be seen that AR in the two biofilters has a higher content of organic matters and microbe than original AR. During the process of leachate treatment, organic content, aerobic and anaerobic bacteria increased with organic load of leachate.

2.2 Treatment of leachate by AR biofilter

The AR biofilter started on 1st December 2006, and the influent and effluent of two-stage biofilter were analyzed periodically for over six months. The influent leachate was taken from a collecting basin in this landfill. The influent leachate is odorous and black with color higher than 1600 (Pt/Co degree). After leachate passed through the AR biofilter, significant reduction in color was observed (lower than 150) and the final effluent was inodorous and pale yellow in color. Regardless of the influent SS concentrations as high as 2324 to 4710 mg/L, the effluent from the AR biofilter was very clear and transparent, with SS lower than 150 mg/L (Fig. 2).

2.2.1 Removal of organic pollutants

The composition and concentrations of organic matters in the influent vary significantly with climate and practical operation of Shanghai Refuse Landfill. The COD con-

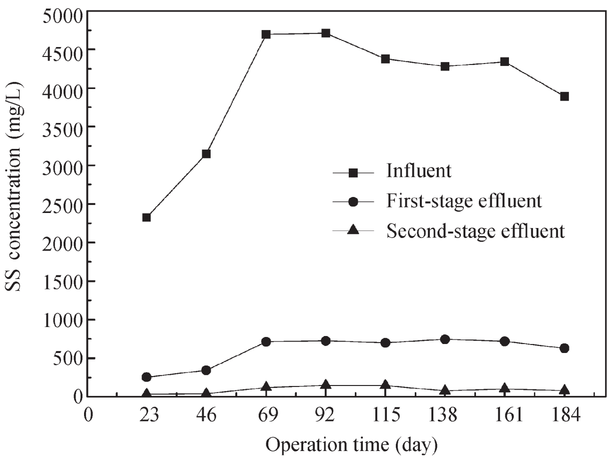

Fig. 2 Solid sludge concentrations of influent and two-stage effluent.

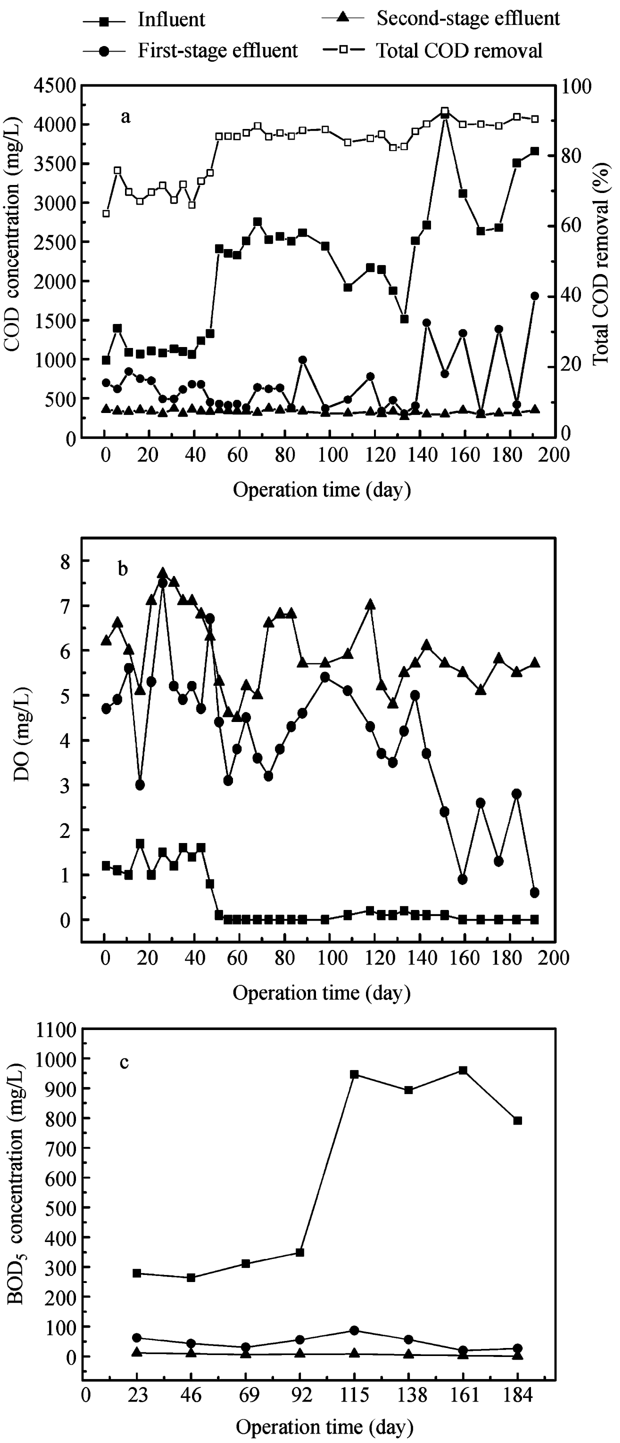

Fig. 3 COD (a), DO (b), and BOD_5 (c) concentrations of influent and two-stage effluent.

Table 1 Analysis results of aged reuse (AR)

Parameter	AR (original)	AR (first stage)	AR (second stage)
pH (± SD, *n* = 3)	7.84 ± 0.01	8.68 ± 0.01	8.39 ± 0.01
Content of organic matters (%) (± SD, *n* = 3)	11.55 ± 0.03	13.60 ± 0.05	12.84 ± 0.03
Aerobic bacteria (CFU/g)	5.80×10^4	6.10×10^7 (winter) 9.10×10^7 (summer)	1.26×10^7 (winter) 8.40×10^7 (summer)
Anaerobic bacteria (CFU/g)	2.00×10^3	1.02×10^6 (winter) 4.40×10^6 (summer)	1.56×10^6 (winter) 2.80×10^6 (summer)

SD: sample standard deviation.

centrations of first-stage effluent decreased to 306–1806 mg/L correspondingly with COD concentrations of influent fluctuating from 986 to 4128 mg/L (Fig. 3a). In spite of the concentrations fluctuation of organic pollutants in the leachate, the final COD concentrations of the second-stage effluent can be reduced to 263–371 mg/L. Overall COD removal by AR biofilter increased gradually from 64% to 93%. With the removal of organic pollutants, the DO value of leachate increased from 0–1.7 mg/L in the influent to 4.5–7.7 mg/L in the effluent, indicating low pollutants content in the final effluent (Fig. 3b).

Even though the BOD_5 concentrations in the influent fluctuating from 264 to 959 mg/L, the effluent concentrations were always below 2–12 mg/L with a removal of 95.8%–99.8%, indicating that AR biofilter is efficient in removing biodegradable organic compounds that are found in the complex landfill leachate (Fig. 3c). On the other side, the BOD_5/COD ratio of the final effluent was lower than 0.03.

2.2.2 Removal of nitrogen

The concentrations of ammonia in the influent and effluent of two-stage AR biofilter are presented in Fig. 4a. The influent ammonia concentrations in the leachate ranged from 538 to 1583 mg/L, as the cases of the corresponding of COD concentrations. After the treatment of first-stage AR biofilter, the ammonia concentrations dropped to 17–774 mg/L with remarkable fluctuation still. The corresponding ammonia concentrations of the final effluent remained almost in the same range (at 2–19 mg/L), with overall removal efficiencies varying from 96.9% to 99.8%.

On the other side, it was found that most NH_4^+-N in the leachate was converted to NO_2^--N in the first-stage AR biofilter (Fig. 4b), and then to NO_3^--N later in the second-stage AR biofilter (Fig. 4c). Figure 4d shows the total nitrogen (TN) concentrations of influent and two-stage effluent. With the TN concentrations of influent ranging from 917 to 1438 mg/L, approximately 49%–63% of the total nitrogen can be removed.

2.3 Determination of bacteria and pH in the leachate

Bacterial quantities of both influent and effluent were also counted by standard methods (APHA, 1998), and the results are shown in Table 2. The microbe in the effluent were so active that about 10^5–10^6 CFU/mL of both aerobic bacteria and anaerobic bacteria were detected while the influent contained less than 10^4 CFU/mL. The pH values decreased from 7.8–8.6 in the influent to 7.1–8.0 in the effluent (Fig. 5).

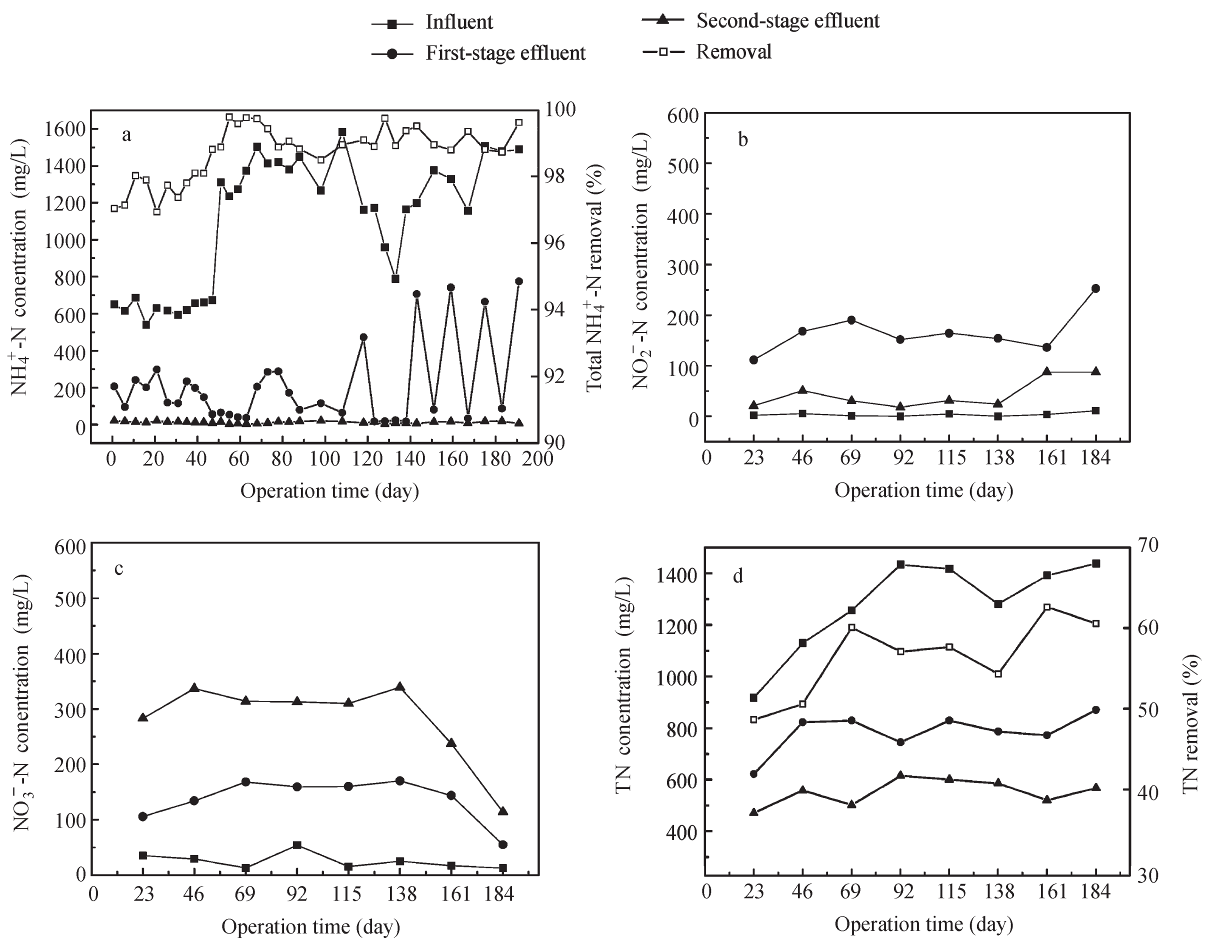

Fig. 4 NH_4^+-N concentrations and total NH_4^+-N removal (a), NO_2^--N concentrations of influent and two-stage effluent (b), NO_3^--N concentrations of influent and two-stage effluent (c), and TN concentrations and TN removal (d).

Table 2 Population of bacteria in the leachate

Population of bacteria	Influent	First-stage effluent	Second-stage effluent
Aerobic bacteria (CFU/mL)	1.00×10^4	4.41×10^6	6.20×10^5
Anaerobic bacteria (CFU/mL)	7.50×10^3	8.20×10^5	5.38×10^6
Escherichia coli (CFU/mL)	3.26×10^6	1.08×10^2	1.38×10^{-1}

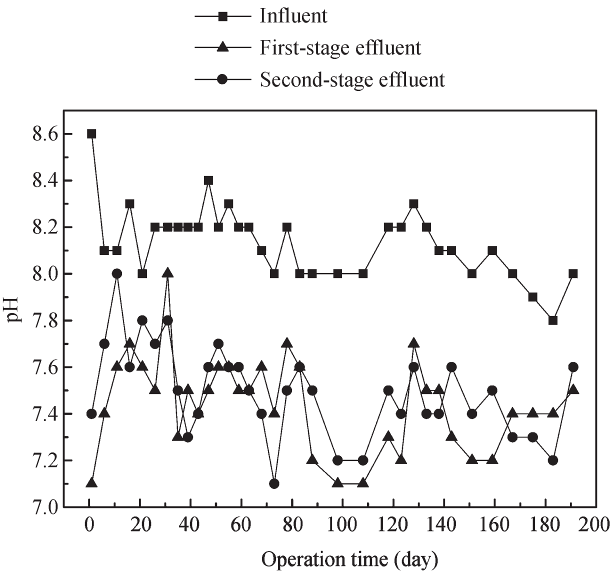

Fig. 5 pH difference between the influent and two-stage effluent.

3 Discussion

Shanghai Refuse Landfill, in which over 25 million tons of aged refuse are there, is the largest municipal landfill in China. Generally, the COD concentrations of raw leachate were above 10,000 mg/L due to large percentage of organics in the landfilled refuse. To mitigate the heavy pollutant impact of leachate on AR biofilter, an pretreatment by covering the leachate regulating container with HDPE membrane was conducted, in which leachate underwent anaerobic self-digestion. It was found that the COD concentrations decreased from 12,000 mg/L to about 2000–4000 mg/L gradually over a period of 100 days. This method could be a practicable and cost-effective technique for it can pretreat high-strength leachate with few operational costs.

The AR biofilter has a similar set-up with low rated trickling filter, which supports attached growth of bacteria by allowing wastewater trickle through the support material due to gravity (Langwaldt and Puhakka, 2000). Trickling filter system has been widely used for the treatment of potable water (Tekerlekopoulou and Vayenas, 2003), dye wastewater (Kornaros and Lyberatos, 2006), municipal wastewater (Elmitwalli et al., 2003; Evans et al., 2004), gold milling effluent (Evangelho et al., 2001), dairy wastewater (Raj and Murthy, 1999a, 1999b), volatile fatty acid (Tsang et al., 2008), waste gas (Tian et al., 2007; Wang et al., 2007), and chromium (Dermou and Vayenas, 2007) etc. This kind of unit is relatively simple and normally produces a consistent effluent quality even with varying influent strength. However, landfill leachate treatment adopting the trickling filter has not been reported yet.

Moreover, differing from the typical support materials used in trickling filters, such as lumps of crushed rocks, slag or pumice, and plastic fills, the AR biofilter adopted aged refuse as the basic media. Furthermore, during the treatment of large amounts of leachate, AR biofilter generated negligible quantity of sludge. Organic pollutants in the leachate were first adsorbed by the AR which acted as microbe carrier, and then were decomposed by great quantity of microorganisms gradually. This process relatively prolonged sludge retention time (SRT), leading to the self-degradation of sludge inside the bed. As a consequence, the AR biofilter does not need secondary sedimentation unit for the sludge like most of the trickling filters have (Wang et al., 2007).

Leachate was treated by the AR biofilter in an open circumstance as temperature was not controlled in our practical operation. Although researchers stated that temperature is a significant factor for ammonia removal (Berge et al., 2007b), operating at different temperature of the AR biofilter did not influence the total ammonia removal. Comparing with the external temperature data got from a local weather bureau nearby (Fig. 6), it was found that the external temperature did not greatly affect the treatment efficiencies of AR biofilter. As can be seen from Fig. 3a, c, and Fig. 4a, the effluent quality in the summer (20–25°C) showed unremarkable improvements with that in the winter (5–10°C). The results were consistent with our laboratory experiment (Zhao et al., 2002). The microbe analysis in Table 1 showed that the populations of both aerobic bacteria and anaerobic bacteria in the winter were almost the same active as (slightly lower than) that in the summer, which could be one of the reasons for the AR biofilter running fluently in low temperature. On the other hand, the height of demonstration AR biofilter is

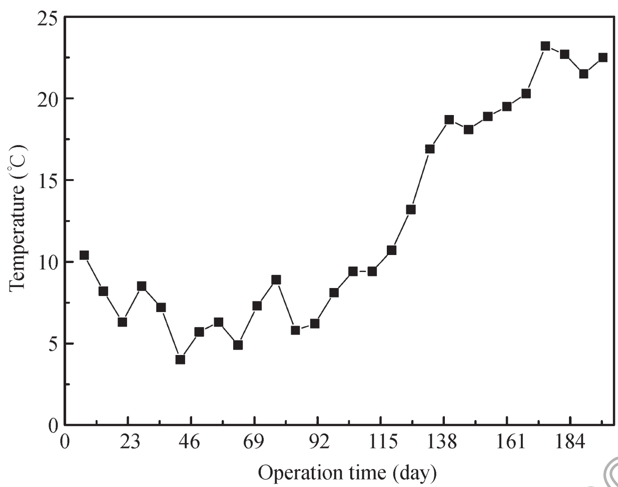

Fig. 6 Linear trend of external temperature.

as high as 3 m, indicating it may keep heat imparted by microorganisms inside the bed successfully.

Understanding microbe removal mechanism of AR biofilter is important. Aerobic and anaerobic bacteria cooperate to decompose pollutants in the leachate. In our previous experiment, the removal efficiencies of AR biofilter dramatically decreased after the AR was disinfected by 20% NaClO (Zhao et al., 2007). Moreover, the anaerobic bacteria in the AR are more active in the two stages than original AR (Table 1). Therefore, it can be concluded that AR biofilter treat landfill leachate biologically with large quantity of microbe.

The AR biofilter had good resistance to pollutant impact in spite of dramatic fluctuation of influent COD concentrations and could remove organic pollutants from leachate very effectively. Although the laboratory experiment results showed the COD concentration in the effluent leachate were always below 300 mg/L (Zhao et al., 2002), the effluent COD concentrations of demonstration AR biofilter were 263–371 mg/L. This means the demonstration biofilter in practice may have a little negative engineering enlargement effect. The BOD_5 results showed the biodegradable organic matters were removed effectively. After the AR biofilter, BOD_5/COD results meant the effluent contained high refractory substances that were difficult to be biodegraded.

As can be seen from Fig. 4a, more than 96.9% of ammonia was removed from leachate by the AR biofilter, followed by an increase of nitrite and nitrate in the effluent (Fig. 4b, c). This means that the AR biofilter has strong nitrification capability for ammonia, leading to high concentrations of nitrite in the first-stage effluents and nitrate in the second-stage effluents. This nitrification process is consistent with the classic two-step route of autotrophic nitrification: ammonia oxidation into nitrite, followed by further oxidation of nitrite into nitrate. As a result, the NO_2^--N concentrations reached the maximum in the first-stage effluent and the NO_3^--N concentrations reached the maximum in the final effluent. The occurrence of nitrification processes reflected the activity of nitrifying microbial populations (Nitrosomonas-like ammonia oxidizers and Nitrospira-related nitrite oxidizers) inside the AR biofilter, as described by Mertoglu et al. (2006) in a bioreactor landfill.

After running for 138 days, a nitrite accumulation occurred in both AR biofilters (Fig. 4b). With respect to the external temperature in Shanghai was nearly 20°C (Fig. 6) at that time and good heat preservation of biofilter mentioned previously, the temperature within the bed maybe exceed 35°C. The high temperature may lead to an immediate nitrite accumulation occurred in the system, as described by Bougard et al. (2006). In a field-scale bioreactor (Berge et al., 2007a), less denitrification occurred when the temperature increased from 22 to 35°C and even 45°C, indicating denitrification may be inhibited by higher temperature. As a result, one potential explanation for the decrease of nitrate toward the end (Fig. 4c) was nitrite accumulation instead of more denitrification.

In wastewater nitrification process, at elevated temperature (> 15°C), ammonium oxidizers have a higher growth rate than nitrite oxidizers (Bougard et al., 2006). Considering the good efficient in preserving heat, the biofilter had a inside temperature always higher than 15°C, even in winter when the external temperature was lower than 5°C (Fig. 6) in Shanghai, as maybe the reason of high nitrite concentrations in the effluent.

The removal of total nitrogen (Fig. 4d) implied that nitrification was not the only nitrogen removal process. Approximately 49%–63% of the total nitrogen was removed, mainly attributing to autotrophic denitrification of nitrate, which was also observed in simulated bioreactor landfills by other researchers (Berge et al., 2007a; Onay and Pohland, 2001). The pH in the influent (average was 8.14) declined with ammonia removal, consistent with the results in a laboratory-scale bioreactor (Berge et al., 2007a), which indicated the dominating ammonia removal mechanism was biotic and most probably due to nitrification. The reason for the relative poor denitrification capacity of the AR biofilter consisted of two parts. One is the shortage of organic carbon source as an electron donor after the first-stage biofilter (BOD_5 lower than 87 mg/L), which may prohibit the denitrification process (Quan et al., 2005). Furthermore, the high DO value of the effluent after first-stage biofilter indicated that excess oxygen may inhibit the growth of denitrifying bacteria (Casasus et al., 2005).

The population of *Escherichia coli* was as high as 3.26×10^6 CFU/mL in the influent. During the leachate treatment, a serial of complex physical, chemical and biologic reaction took place within the AR biofilter, which could kill the *Escherichia coli*, leading to a low concentration of the *Escherichia coli* in the effluent (less than 10^{-1} CFU/mL).

During the operation of 191 days, the AR biofilter ran stably and continuously. Occasionally, the surface of first-stage biofilter may be covered by black silt due to the high SS in the influent (Fig. 2), which may lead to the accumulation of leachate on the surface. This problem can be solved by raking the surface from time to time.

Through the excavation in landfill, some stuff such as plastic materials and glass bottles could be recycled and reused. On the other hand, AR could be used as biofilter materials for the effective treatment of landfill leachate. Landfilling compartments could be relandfilled solid wastes into, leading to a longer service time of the landfill.

4 Conclusions

A 100-m^3 leachate/day demonstration AR biofilter has been successfully operated at the Shanghai Refuse Landfill. During the operation process of AR biofilter, the COD concentrations decreased from 986–4128 mg/L to 263–371 mg/L with BOD_5/COD ratio lower than 0.03, and ammonia concentrations reduced from 538–1583 mg/L to 2–19 mg/L. This treatment process is very effective, which takes advantages of the variety and high population microorganisms present in the AR excavated from landfill to decompose the recalcitrant pollutants in landfill leachate.

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