

A laboratory simulation of *in situ* leachate treatment in semi-aerobic bioreactor landfill

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Abstract

In this study, two laboratory-scale simulated landfill bioreactors were established, of which Reactor A was operated only with leachate recirculation and served as the control, and Reactor B was operated as semi-aerobic bioreactor landfill with leachate recirculation. *In situ* leachate treatment and accelerating organic decomposition in semi-aerobic bioreactor landfill was investigated. The results indicated that the introduction of air into the landfill was favourable for optimising the micro-organism growth environment and accelerating the degradation of organic matter. It can be seen clearly from the results that $\text{NH}_4^+\text{-N}$ can be removed *in situ* in the semi-aerobic bioreactor landfill with leachate recirculation. Moreover, semi-aerobic bioreactor landfill showed lower emissions for leachate than those in leachate from anaerobic landfill, with low concentrations of COD, VFA, $\text{NH}_4^+\text{-N}$ and TKN, and which saved the disposing process of the discharged leachate. The three-dimensional excitation-emission matrix fluorescence spectroscopy (EEMs) of dissolved organic matter (DOM) in Reactor B changed greatly, and fluorescence peak changed from protein-like fluorescence at Day 60 to humic-like fluorescence at Day 95 and 250, while in Reactor A, fluorescence peak of DOM was always protein-like fluorescence. The comparison of the EEMs indicated that the semi-aerobic landfill accelerated the organic decomposition.

Keywords: semi-aerobic landfill, bioreactor landfill, three-dimensional excitation-emission matrix fluorescence spectroscopy (EEMs), *in situ* leachate treatment

Introduction

In 2002, approximately 94% of the municipal solid waste (MSW) generated in China was disposed of in landfills (Wang et al., 2006). However, anaerobic degradation of solid waste results in the production of leachate and landfill gas for a very long time in a conventional landfill. Leachate and landfill gas are the potential pollution sources for the surrounding environment, and the long-term environmental impacts will last for several decades (Cossu et al., 2003; Bilgili et al., 2006). Therefore, there has been increased emphasis on the operation of landfills as bioreactors to enhance decomposition of solid waste, provide a reduction in landfill emissions over a relatively short time, and dispose leachate *in situ* (Price et al., 2003; Mehta et al., 2002; Pohland et al., 2000; Reinhart et al., 2002; Reinhart, 1996; Pohland et al., 1994; Townsend et al., 1996; Chan et al., 2002; Demir et al., 2004; Bilgili et al., 2004; Mark and Cristina; 2006). However, ammonia nitrogen typically accumulates because ammonia is stable under anaerobic conditions. Thus, higher concentrations of ammonia than those found in leachate from conventional landfills last long even after the organic fraction of the waste is stabilised (Burton and Watson-Craik, 1998; Onay and Pohland, 1998; Price et al., 2003). That is the reason why ammonia removal is an important aspect of long-term landfill pollution control.

However, *ex situ* leachate treatment of high concentration COD and $\text{NH}_3\text{-N}$ can be costly (Ferhan and Aktas 2000;

Maree et al., 2004). Recently, *in situ* biological nitrogen removal for bioreactor landfills attracted more and more attention of the researchers. Air addition has recently been practised at a number of pilot-scale and field-scale landfills worldwide, where it was found that the organic fraction of the waste decomposed far more rapidly under aerobic conditions than under anaerobic conditions, and ammonia was removed *in situ* by nitrification and denitrification in aerobic landfills (Berge et al., 2006; Onay et al., 2001; Reinhart et al., 2002; Read et al., 2001; Das et al., 2002; Themelis et al., 2001; Borglin et al., 2004; Boni et al., 1997). However, there are some disadvantages for the aerobic bioreactor landfills, such as needing forced ventilation systems, complex operation and management, and large energy consumption. Semi-aerobic landfills use natural ventilation instead of mechanical ventilation for oxygen supply, and which create an aerobic region in the landfill. Semi-aerobic landfill system is propitious to the simultaneously occurring nitrification and denitrification and thus *in situ* disposes the leachate effectively and accelerates the stabilisation of the waste (Theng et al., 2005). In China, only a few researchers studied the semi-aerobic landfill technology, but no systematic and comprehensive studies have been conducted (Wang et al., 2006).

Moreover, fluorescence excitation-emission matrix spectroscopy (EEMs) provides much more detailed information about fluorescence properties of the organic matter that may reveal important information about its composition and biogeochemical cycling (Burdige, et al., 2004). Fluorescence spectroscopy has high sensitivity and specificity, and has already been used in natural water quality monitoring. Rapid analyses are now possible using fluorescence spectrophotometers such that the EEMs

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can be generated in approximately 1 min (Baker, 2002; Baker and Curry, 2004). Thus, fluorescence analyses may be providing a rapid method to characterise organic matter and to fingerprint organic pollutants in the leachate.

The main objective of the research was to investigate *in situ* leachate treatment and accelerating organic decomposition and stabilisation of solid waste in semi-aerobic landfill by chemical analysis and fluorescence analyses.

Materials and methods

Experimental equipments

Two simulated reactors, made of polymethylmethacrylate (PMMA) of 1.5 cm thickness, with a diameter of 20 cm and height 1.2 m, providing an effective volume of 31.4 l, were used in this experiment. Both of the reactors were wrapped with 1 cm thick polyurethane polyfoam which acted as a heat-insulating material to prevent temperature redistribution between the reactors and the surrounding environment. Prior to filling, a 10 cm thickness of gravel was placed at the bottom of each landfill reactor to retain refuse and stop small particles from leaching out. Then about 17.3 kg of pretreated MSW mixture was filled into each landfill reactor. Finally, the waste mixture was covered with a 10 cm depth of pea gravel and a water distributor was installed at the upper part of every reactor. Three sample collection ports of solid wastes were set in the middle of the reactors; the leachate outlets, which were connected to the leachate recirculation tanks, were under the reactors. Besides, the vent-pipe was installed under the reactor B (Fig. 1).

The temperature in a Chinese landfill ranges from 22 to 52°C, and is 27 to 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 (He et al., 2006). Thus, two simulated reactors were both placed in a thermostatic chamber and the temperature was kept at 33±2°C. After filling, Reactor A was sealed with silicone caulk and monitored for leaks. Then, approximately 3 l deionised water was added to the reactors in order to produce about 0.5 l of leachate at the start of the experiment.

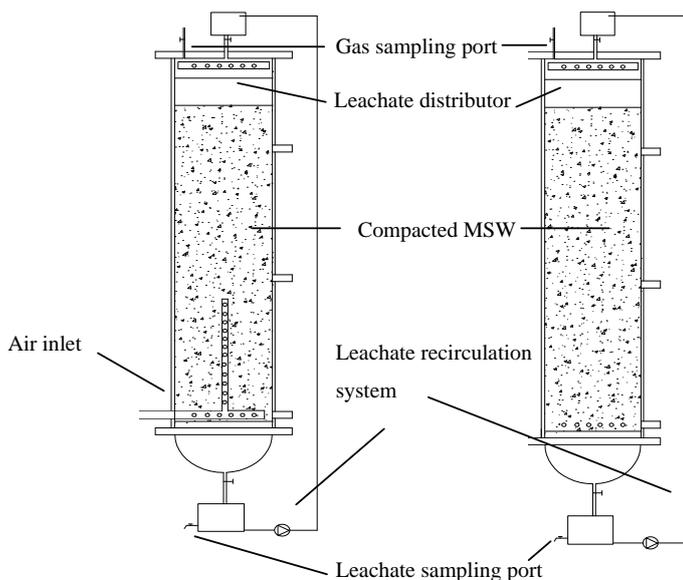


Figure 1

Schematic of simulated anaerobic and semi-aerobic reactors

Experimental materials

Fresh waste was obtained from Asuwei Sanitary Landfill Site, located in Beijing, China. Plastic bags and large inorganic waste were removed by a first sorting step and further sorting was conducted in the laboratory. All of the waste was then shredded and mixed to avoid leachate preferential flow in simulated landfill reactors. The waste composition was as follows (by weight): Kitchen waste, 70.3±8.3%; paper, 10.2±2.1%; plastic, 8.1±1.9%; fabric, 1.2±0.6%; metal, 0.2±0.1%; and others, 1.0±0.2%. The relatively high proportion of kitchen waste or organic waste is considered to be a characteristic of MSW in China (He et al., 2005, 2006a; He and Shen, 2006; Wang et al., 2006), as well as several other developing countries (Sponza and Agdag, 2004; San and Onay, 2001).

Reactor operation

Reactor A served as a control; leachate was drained to a recirculation tank from Reactor A, and recycled by peristaltic pump every 2 d. Reactor B was operated similarly as Reactor A, but after Day 60, the valve of the vent-pipe was opened in order to simulate the semi-aerobic bioreactor landfill. Water was sprayed into the top of the reactors at about 300 ml/6d to increase the moisture content of the waste. The daily load of recycled leachate to reactors did not exceed 4 l in order to avoid flooding of reactors.

Sampling and analytical methods

Leachate samples were collected from the recirculation tank every 2 d and analysed for chemical oxygen demand (COD), biological oxygen demand (BOD₅), pH, ammonia nitrogen (NH₄⁺-N), NO₃⁻-N, NO₂⁻-N, Total Kjeldahl nitrogen (TKN) according to the *Standard Methods* (EPA of China, 1989). The pH value was measured by a Sartorius digital pH meter with PY-AS1 electrode (PB-10, Sartorius Inc., Germany). The ORP was measured by a Thermo Orion model 250A4 ORP meter with a 9179BN electrode (Thermo Orion Inc., USA). The concentration of the VFA was determined by gas chromatography (GC-6890N, Agilent Inc., USA). The gas content in biogas samples was analysed using a second gas chromatograph (HP5890, HP Inc., USA).

The leachate samples were centrifuged with a rotating speed of 12 000 r·min⁻¹ for 20 min under 4°C, and filtrated through glass-fibre membrane (0.45 µm) to remove the suspended matters that may react with DOM. The organic matter in the supernatant was dissolved organic matter (DOM). The total organic carbon (TOC) of the DOM was determined by TOC analyser (SHIMADZU TOC-5000). Methods described by Christensen (1998) and Ma et al. (2001) were adopted herein for dissolved organic matter (DOM) fractionation in leachate. The DOM in the leachate was fractionated into humic acid (HA), fulvic acid (FA) and hydrophilic (HyI) fractions. Amberlite XAD-8 resin was obtained from Rohm and Haas Co. and 732 cation exchange resins were obtained from Beijing Huideyi Co. Ltd. (Beijing, China). XAD-8 and 732 resins were cleaned by the method described in the literature (He et al., 2006b). The concentrations of HA, FA and HyI fractions were determined through measuring dissolved organic carbon (DOC) concentration of the fractionated sub-samples.

The EEMs was measured by fluorescence spectrometer (Perkin Elmer Luminescence Spectrometer LS50B). Each EEM was generated by scanning excitation wavelengths from 200 to 480 nm at 10 nm steps, and detecting the emitted

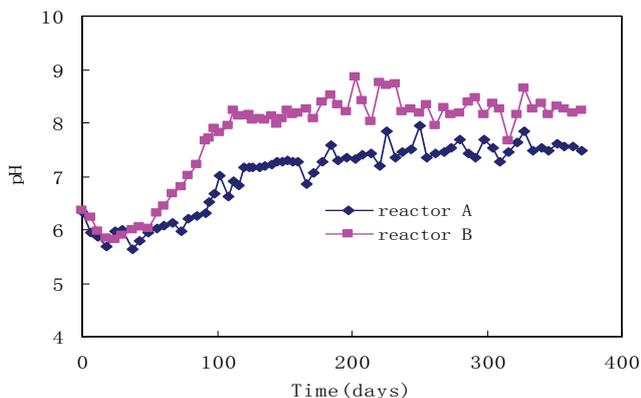


Figure 2

The change of pH in semi-aerobic and anaerobic reactors

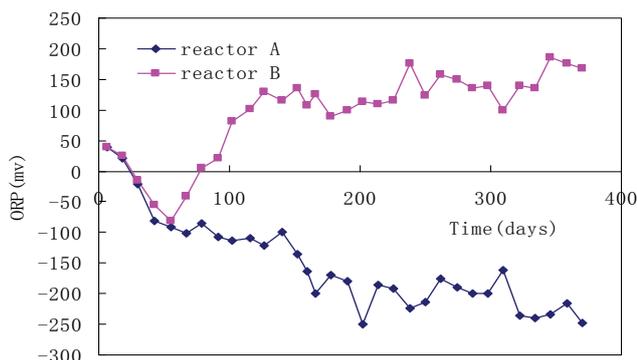


Figure 3

The change of ORP in semi-aerobic and anaerobic reactors

fluorescence between 200 and 600 nm at 10 nm steps. Scan speed was 1 200 nm·min⁻¹, permitting collection of a complete EEM in ~60 s. EEM spectra are illustrated as the elliptical shape of contours. The X-axis represents the emission spectra from 200 to 600 nm, whereas the Y-axis is the excitation wavelength from 200 to 480 nm. Contour lines, as the 3rd dimension, are shown for each EEM spectra to represent the fluorescence intensity.

Results and discussions

pH and ORP

Figure 2 shows the change of pH over time. The pH values were in the range of 5.5 to 6.5 in the first 60 d of degradation in two reactors. After Day 60, pH values began to increase and reached 8 after Day 108 in semi-aerobic reactors. In the subsequent days, no considerable variation was observed for pH in leachate from semi-aerobic reactor and pH values remained about 8. Different from semi-aerobic reactor, on Day 108, the pH was 6.6 in anaerobic reactor, and reached 7.5 after Day 180. These results indicated that the semi-aerobic reactor reaches optimal pH value much faster than that the anaerobic reactor, showing the rapid degradation of solid wastes in semi-aerobic condition. These results are in accordance with previous studies (Bilgili et al., 2006; Nakasaki et al., 1993; Cossu et al., 2003).

Oxidation-reduction potential (ORP) is used to monitor chemical reactions, to quantify ion activity, or to determine the oxidising or reducing properties of a solution. It also provides information about the biological processes occurring under aerobic and anaerobic conditions. The ORP within a landfill deter-

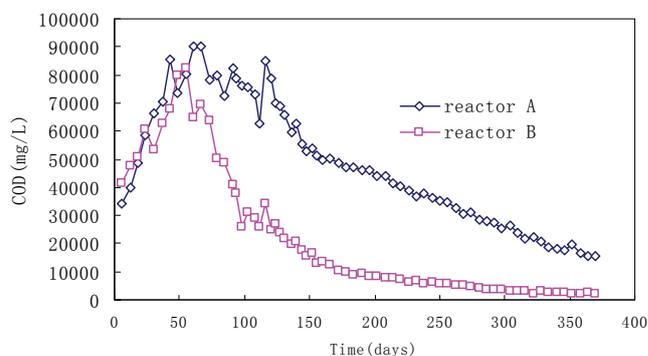


Figure 4

Changes of COD concentrations in semi-aerobic and anaerobic reactors

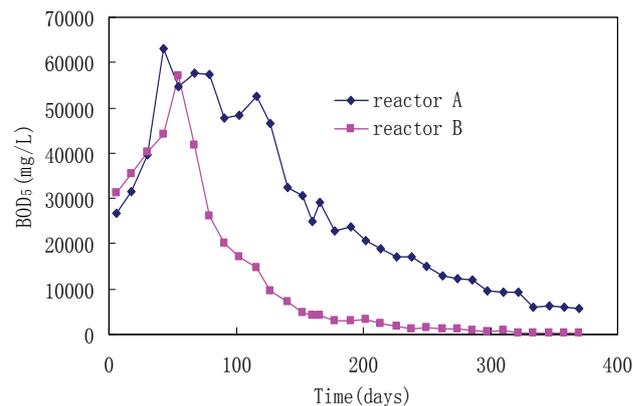


Figure 5

Changes of BOD₅ concentrations in semi-aerobic and anaerobic reactors

mines the mechanism of waste degradation. Bilgili et al. (2006) reported that high ORP (aerobic conditions) causes accelerated degradation of waste. The results of ORP are plotted in Fig.3. In the beginning of the 60 d period, the ORP values were the same in the two reactors. After Day 60, the ORP values was increased in semi-aerobic reactor and reached 100 mV on Day 116. Oppositely, ORP values decreased below -100 mV in the anaerobic reactor after 90 d, which showed that the degradation was changing from the acidogenic phase to the methanogenic phase after the consumption of the available oxygen in the anaerobic reactor. Some researchers found that there is an optimum ORP requirement for methanogenesis, which generally ranges from -100 to -300 mV (Bilgili et al., 2006).

Organics and gas composition

COD data for semi-aerobic and anaerobic reactors are presented in Fig. 4. The initial COD concentrations were around 40 000 mg/l in leachate for Reactor A and B. COD concentrations increased rapidly after the experiment commenced, and reached the maximum values of 82 600 and 90 200 mg/l for Reactor A and B after 65 and 55 days, respectively. After reaching to maximum values, COD concentrations began to decrease rapidly for Reactor B, and the decreasing rate of concentration from Reactor B was greatly faster than that from Reactor A. The final concentrations measured in Reactor A and B on Day 370 was 15 600 and 2 270 mg/l, respectively.

Changes of BOD₅ concentrations are presented in Fig. 5. The BOD₅ results showed similar trend with COD. The final concentrations measured in Reactor A and B on Day 370 were

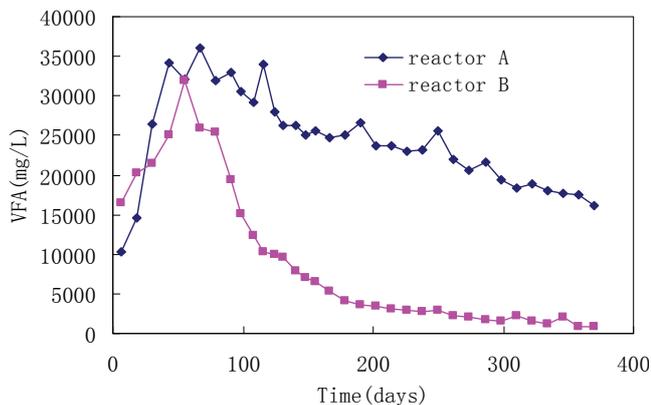


Figure 6
Changes of VFA concentrations in semi-aerobic and anaerobic reactors

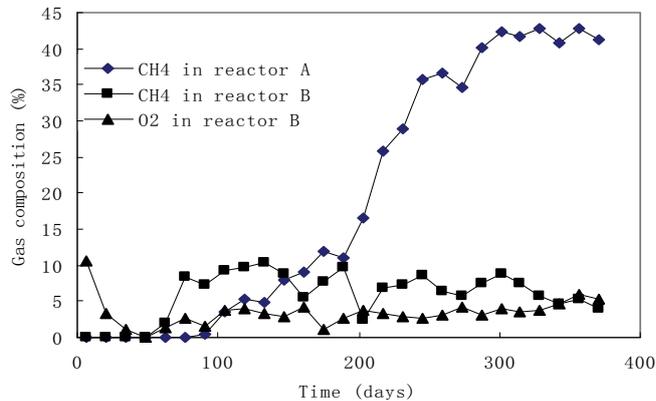


Figure 8
Time evolution of gas compositions in semi-aerobic and anaerobic reactors

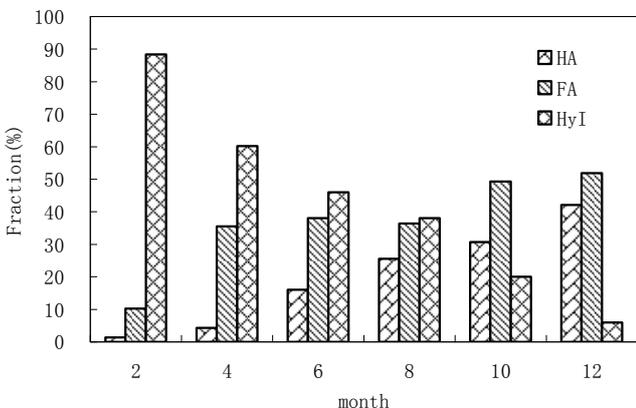
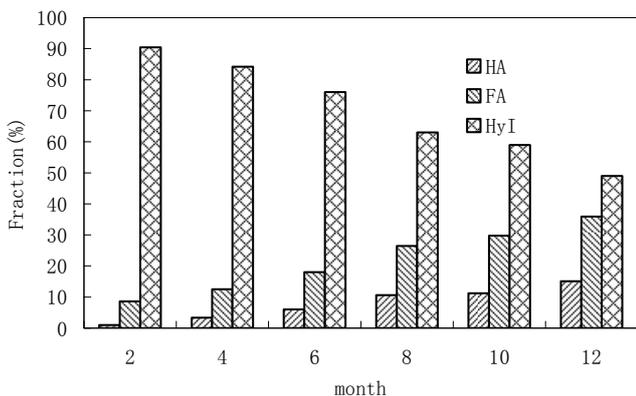


Figure 7
Changes of HA, FA and HyI in leachate samples of semi-aerobic and anaerobic reactors

5 600 and 360 mg/l, respectively, and correspondingly, the ratio of BOD/COD was decreased from 0.78 and 0.76 at the beginning of operation to 0.36 and 0.16, respectively. The BOD/COD ratio indicates the amount of biodegradable compounds in the leachate. The BOD/COD ratio of Reactor B was lower than that of Reactor A; this may be as a result of accelerating biodegradable organic degradation under semi-aerobic condition. These results are in accordance with previous studies (Bilgili et al., 2006; Cossu et al., 2003). Their studies show that the COD values of leachate from aerobic reactors were lower than those in the leachate from the anaerobic reactor.

Volatile fatty acids (VFA) are the most important inter-

mediates in the anaerobic digestion process. Moreover, since VFA accumulation may lead to process failure due to the pH drop they induce (Anderson and Yang, 1992), VFA concentrations have been monitored for a long time as process performance indicators. The variation of VFA in leachate from semi-aerobic and anaerobic reactors is presented in Fig. 6. As illustrated in the figure, VFA concentration accumulated during the beginning of 60 d in Reactor B and reached a maximum value of 31 900 on Day 60, and then decreased quickly. However, VFA concentration accumulated and increased to a maximum value of 34 000 on Day 70 in Reactor A, and then decreased slowly.

The variation of HA, FA and HyI in leachate samples is presented in Fig. 7. The results showed that the DOM in leachate was mainly composed of an HyI fraction in Reactors A and B in the first 60 d. After semi-aerobic operation for Reactor B, the HyI fraction considerably decreased; correspondingly, the HA and FA fractions increased quickly, while the variation of DOM compositions was not obvious in Reactor A. All of these results show that the residual organic comprised non-biodegradable large-molecule compounds in the leachate from Reactor B, such as humic acid, which was difficult to biodegrade (Zouboulis et al., 2003).

The methane appeared in the gas phase of Reactor A and Reactor B since Day 100 and 65, respectively (Fig. 8). The volume fraction of methane increased slowly and reached 40% after 300 d in Reactor A. Different from Reactor A in which methanogenesis was largely inhibited by VFA accumulation and low pH, methane started to appear since the first week in the Reactor B. However, the volume fraction of methane never exceeded 10%, and the corresponding O₂ level was approximately 2%.

These results indicated that semi-aerobic landfill can optimise micro-organism growth environments, and have a positive effect on the balanced growth of the acid-production phase and methane production (Dong et al., 2007), and accelerate organic decomposition. However, the activity of methanogen was inhibited in the anaerobic landfill reactor due to the accumulation of organic acid during the acid-production phase, and organic matter degradation in the leachate was slowed down consequently.

Nitrogenous compounds

The data of NH₄⁺-N and TKN in semi-aerobic and anaerobic reactors are given in Figs. 9 and 10, respectively. The NH₄⁺-N concentrations in the two reactors increased greatly in the initial stage and were kept constant after 40 d. The maximum

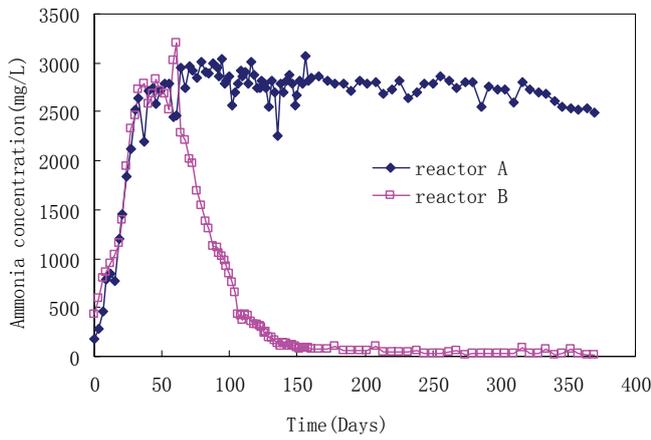


Figure 9
Changes of $\text{NH}_4\text{-N}$ concentrations in semi-aerobic and anaerobic reactors

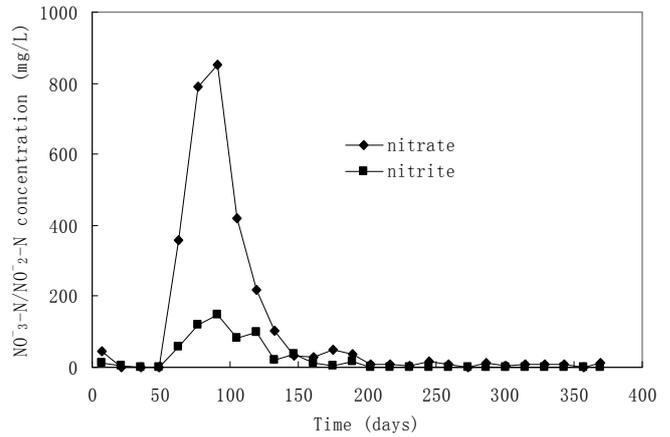


Figure 10
Changes of TKN concentrations in semi-aerobic and anaerobic reactors

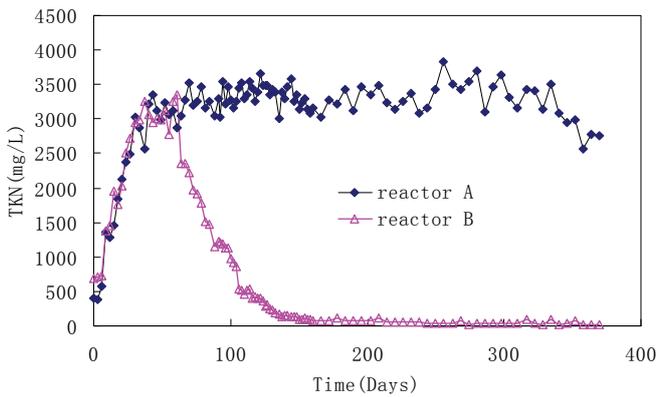


Figure 11
Changes of $\text{NO}_3\text{-N}/\text{NO}_2\text{-N}$ concentrations in semi-aerobic and anaerobic reactors

$\text{NH}_4\text{-N}$ and TKN concentrations were measured to be 3 198 and 3 345 mg/l for semi-aerobic reactor, and to be 2 950 and 3 340 mg/l for anaerobic reactor, respectively. After 60 d the $\text{NH}_4\text{-N}$ concentrations in leachate decreased quickly for Reactor B due to semi-aerobic operation. Moreover, the $\text{NO}_3\text{-N}$ and $\text{NO}_2\text{-N}$ concentrations sharply increased, and then decreased over the subsequent period (Fig. 11). The $\text{NO}_3\text{-N}$ and $\text{NO}_2\text{-N}$ was not detected in Reactor A. The final $\text{NH}_4\text{-N}$ concentrations in Reactors A and B were 2 490 and 22 mg/l on Day 370, respectively. Differences between semi-aerobic and anaerobic reactors indicate the nitrifying effect. The results suggested that semi-aerobic operation is an effective way to enhance biological nitrification and remove $\text{NH}_4\text{-N}$. The reactor was developed to include aerobic, anoxic and anaerobic zone under semi-aerobic condition. The sources of carbon and nitrate necessary for denitrification could be supplied by utilising leachate recirculation to carry the residual C and N from the anaerobic zone into the aerobic zone, and subsequently to the anoxic zone at the top of the system. Accordingly, ammonia nitrogen could be removed *in situ* (Dong et al., 2007).

Most of the nitrogen in landfill bioreactors is $\text{NH}_4\text{-N}$ and is produced from the degradation of proteins and amino acids (Price et al., 2003). Some researchers reported that ammonia was the most significant long-term component of leachate (Christensen et al., 1998), because ammonia is stable under anaerobic conditions. In general, there should be no apparent increase or decrease in the concentration of all nitrogen groups during the

anaerobic degradation of solid waste (Bilgili et al., 2006). Thus, the same trend of TKN concentrations was observed during the study.

As illustrated, it can be seen clearly that $\text{NH}_4\text{-N}$ could be *in situ* removed by semi-aerobic operation with leachate recirculation, which could avoid the disposing process of the discharged leachate. Moreover, during the *in situ* $\text{NH}_4\text{-N}$ removal process, the biodegradable organic matters could be used as carbon source of denitrification in semi-aerobic reactor and thus avoided the inadequate of carbon sources at the late stage of the recirculated landfill.

EEMs of the leachate

In this study, we applied three-dimensional fluorescence EEMs spectra for characterising the leachate samples. Each EEM provided spectral information about the organic matter compositions of leachate samples. Fluorescence spectra such as those we have obtained contains a number of distinct peaks that are generally ascribed to either humic-like or protein-like fluorescence, and at least six peaks were readily identified from EEM fluorescence spectra of leachate DOM (see Fig. 12). The first main peak was a distinctive and intense fluorescence peak at excitation and emission wavelengths (Ex/Em) of 210-230 nm/340-360 nm (peak A), which is identical in location to the diagnostic fluorescence centre observed previously (Baker and Curry, 2004). Moreover, there were three peaks that were identified at Ex/Em of 210-230/300-320 nm, 270-280/300-320 nm and 270-280/340-380 nm, marked by peak B, peak C and peak D, respectively. The four peaks have been ascribed as protein-like peaks, in which the fluorescence is associated with the tryptophan and tyrosine (Baker, 2001; Chen et al., 2003; Yamashita and Tanoue, 2003; Baker and Inverarity, 2004). A fifth peak was located around Ex/Em of 310-340/420-440 nm (Peak E), which is attributed to aromatic and aliphatic groups in the DOM fraction and commonly labeled as fulvic-like (Coble, 1996). In these leachates we also observed a 6th peak at Ex/Em of 220 to 240/420 to 440 nm (Peak F), a poorly understood fluorescent centre widely attributed to a component of the humic fraction (Burdige et al., 2004; Coble, 1996; Yan et al., 2000).

All of the fluorescence peaks of the leachate in the Reactor A on Day 60, 95 and 250 were protein-like fluorescence (some examples are shown as contour maps in Fig. 12a, b, c), which are attributed to protein tryptophan and tyrosine (Burdige et al., 2004; Chen et al., 2003; Lawrence et al., 1999; Wu and Tanoue,

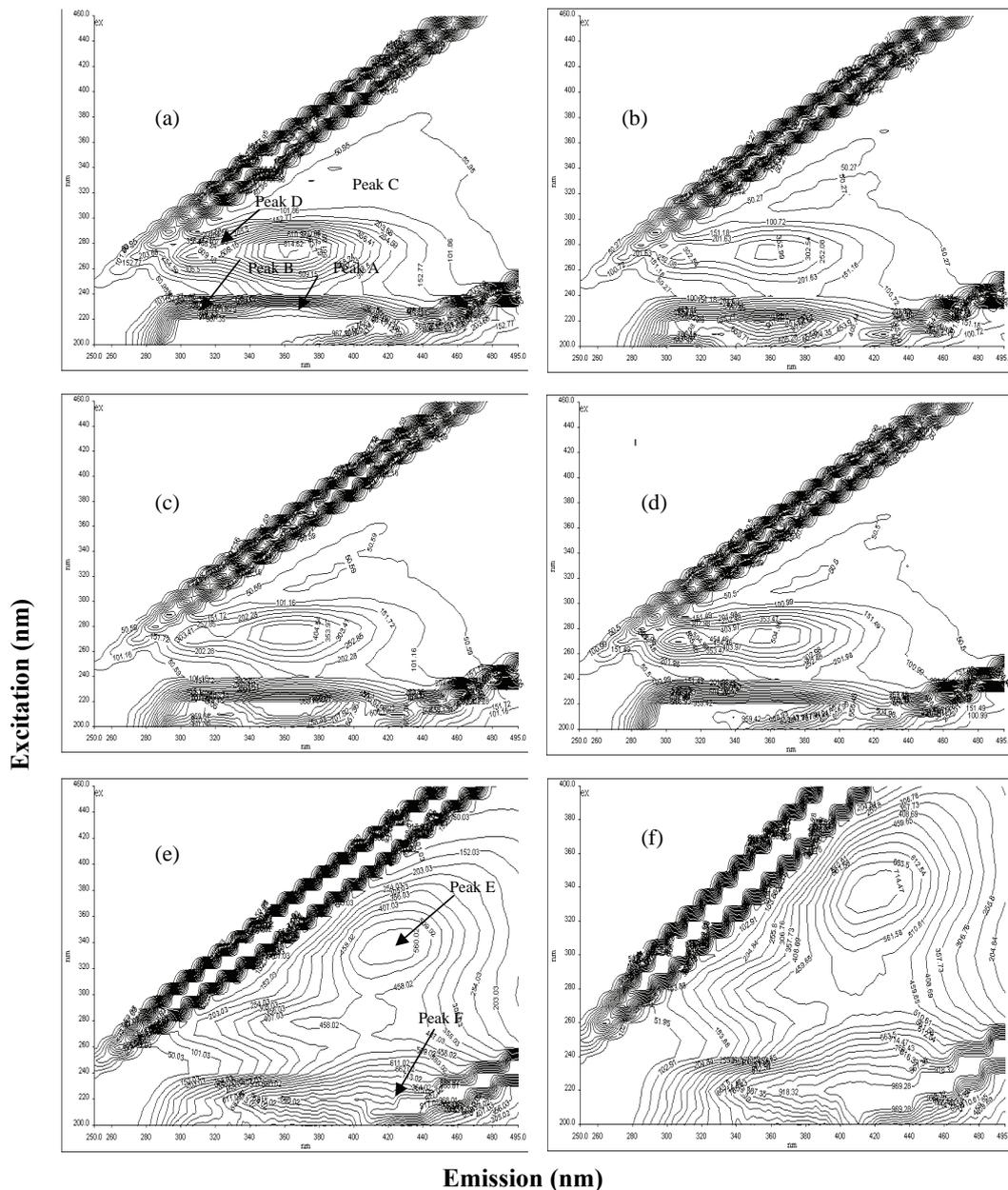


Figure 12
Fluorescence EEM contour plots for DOM of the leachate in Reactors A and B

2001). Peak locations of the three leachate fluorescence spectra were slightly different along with landfill time. The locations of these peaks for the leachate on Day 250 were red shifted to longer wavelengths compared to those of the leachate on day 60 and 95. Such differences implied that the fluorophores presented in the three samples from Reactor A had a little change with landfill time.

However, in the Reactor B, the variation of the fluorescence peaks in the leachate was different from Reactor A. The fluorescence peaks of the leachate was protein-like fluorescence in the first 60 d, and then changed to Peaks E and F (Fig. 12d) on Day 95 and 250, which are attributed to aromatic and aliphatic groups in the DOM fraction and commonly labelled as humic-like and fulvic-like, and related to the carbonyl and carboxyl in the humus (Wu and Tanoue, 2001). The locations of peaks E and F for the leachate on Day 250 were red shifted to longer wavelengths compared to those of the leachate on Day 95 and the

intensity of the fluorescence peaks tend to increase. The change of peak locations and fluorescence intensity may be ascribed to an increase of molecular size, aromatic polycondensation, level of conjugated chromophores and humification degree of organic matter as the landfill time increases. The result was in accordance with previous studies (Senesi and Miano, 1991; Wei et al., 2007).

Some reports presented that most of the organic matters were biodegradable in the leachate at the beginning of landfill, and then became nonbiodegradable (such as humic acid) at the late stage of landfill (Zouboulis et al., 2003). The EEMs of leachate indicated that organic matters were humic-like and fulvic-like in the leachate of Reactor B. Furthermore, the fluorescence results suggest a larger humification degree of leachate in Reactor B with respect to the corresponding leachate in Reactor A. This is also evidence to suggest that semi-aerobic operation stimulates the organic decomposition.

Conclusions

The results indicated that the semi-aerobic landfill could separate the acid formation phase and the methane fermentation phase, optimise the micro-organism growth environment and accelerate the degradation of organic matter. Moreover, it showed lower emissions for leachate than those in leachate from anaerobic landfill, with low concentrations of COD, BOD₅, VFA, ammonia and TKN. *In situ* removal of NH₄⁺-N and organic matter in leachate could be carried out in a recycled semi-aerobic landfill and therefore the disposal process of the discharged leachate is avoided. Compared with traditional anaerobic and aerobic bioreactor landfill operations, the introduction of air into the landfills by natural ventilation induced a rapid and marked oxidation of organic matter and nitrogen. The positive effect of leachate recirculation was seen more clearly in a semi-aerobic bioreactor landfill operation than in the anaerobic bioreactor landfill.

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