# Landfill leachate treatment by MBR: Performance and molecular weight distribution of organic contaminant

CHEN Shaohua & LIU Junxin

Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China
Correspondence should be addressed to Liu Junxin (email: jxliu@rcees.ac.cn)

**Abstract** A membrane bioreactor (MBR) with an air-lift bioreactor and gravity flow is applied to treating landfill leachate. More than 99% of $BOD_5$ (biochemical oxygen demand for five days) removal efficiency is achieved with less than 35 mg/L of $BOD_5$ in the effluent at less than 1.71 kg $BOD_5/m^3 \cdot d$ of $BOD_5$ loading rate. When DO (dissolved oxygen) is maintained at the range of 2.3—2.8 mg/L and the loading rate of $NH_4^+$-N (ammonium nitrogen) is kept at 0.16—0.24 kg $NH_4^+$-N/$m^3 \cdot d$, the $NH_4^+$-N in the effluent is less than 15 mg/L. However, compared with high removal rates of $BOD_5$ and $NH_4^+$-N, the removal efficiency of soluble chemical oxygen demand (SCOD) varies between 70% and 96%. The investigation of molecular weight (MW) distribution has been carried out by the gel permeation chromatography (GPC) so as to understand the fate of organic matters in the MBR treating of landfill leachate. Results indicate that organic matters of the landfill leachate are composed of a high MW fraction (MW of the peak, $MW$p = 11480—13182 Da) and a low MW fraction ($MW$p = 158—275 Da). The high MW fraction is not biodegradable, but can be decreased with microfiltration membrane. The most of the low MW fraction is biodegradable, but the residue of the low MW fraction is able to permeate through the membrane, thus resulting in high SCOD in the effluent of the MBR.

Keywords: **landfill leachate, membrane bioreactor, molecular weight distribution, wastewater treatment.**

Landfilling is the most popular way for municipal solid waste (MSW) disposal and has been widely applied in the world. In 2003, about 148 million tons of MSW was disposed of by sanitary landfilling in China[1]. Leachate produced from the process of landfilling, which contains a large amount of soluble or suspended organic matters, $NH_4^+$-N and inorganic ions, may cause nuisance to adjacent communities and contribute severe environmental hazards when it is not properly collected, treated and safely disposed[2]. Hence, the treatment of leachate is one of the key factors to manage the landfill.

Biological methods, e.g. aerobic and anaerobic techniques, have been used to treat leachates during the last few decades. Anaerobic processes have been shown to be efficient in the treatment of the young leachates with high $BOD_5$[3−5], while activated sludge systems and aerated lagoon systems are extensively used for leachate treatment[6]. Extended aeration of activated sludge with a relatively long hydraulic retention time (3 to 10 d) achieved good results for C and N removal[6]. The treatment of leachates by on-site aerated lagoon plants in Britain and Ireland showed that the effluent $BOD_5$ was rarely over 50 mg/L and more than 97% of COD removal was achieved, together with excellent removal of ammonia, iron, manganese and zinc[7]. It has been proved that the sequencing batch reactor (SBR) is a reliable method for treating landfill leachates[8,9].

It is well known that biological techniques treating landfill leachates are successful in the removal of BOD[7−9] and ammonia[10−12]. However, COD removal is considerably more challenging, because of removal efficiency varying from 20% to more than 90% depending on characteristics of leachate, types and operational facets of process[11,13,14]. In order to meet the more stringent disposal regulations, the processes for landfill leachate treatment currently used are the combination of biological and physical and/or chemical treatment technologies. Generally, a biological technique is firstly applied to removing ammonia, COD and BOD, followed by an additional physicochemical treatment to remove non-biodegradable organic components[15−19].

Recently, to MBRs more attention is paid in landfill leachates treatment owing to their efficiency and small foot-print[15,16,19−24]. The performance of some MBRs for treating landfill leachate is listed in Table 1. Compared with less than 0.25 kgCOD/$m^3 \cdot d$ of conventional activated sludge processes, these MBRs had higher loading rates (0.75-9.0 kgCOD/$m^3 \cdot d$) and achieved more than 94% of $BOD_5$ removal at shorter hydraulic retention time (HRT)[22]. However, alike conventional activated sludge processes, high COD concentration

Table 1   Performance of MBR treating landfill leachate

| Scale | HRT (h) | COD | | | BOD$_5$ | | | NH$_4^+$-N | | | Ref. |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | in (mg/L) | out (mg/L) | removal (%) | in (mg/L) | out (mg/L) | removal (%) | in (mg/L) | out (mg/L) | removal (%) | |
| Full | 96 | 3000 | – | – | <0.1[c)] | – | – | 1200 | 29[a)] | 96[b)] | [15] |
| Full | – | 400－1500 | 211－856 | – | 100－500 | 4.3－29 | – | 200－1400 | 100－408 | – | [16] |
| Lab | 24 | 8000－9000 | 1800－2400 | – | 0.40－0.45[c)] | 60－100 | – | 340－360[d)] | 120－150 | | [19] |
| Lab | 24 | 1800 | – | 31.3 | 267.5 | – | 98 | 114.8 | – | 66 | [21] |

a) Inorganic nitrogen; b) total nitrogen removal; c) BOD$_5$/COD; d) after ammonia stripping.

was still found in effluents of these MBRs (Table 1) due to a certain amount of refractory compounds presenting in landfill leachate[17,19,22]. To make this phenomenon clear, a thorough analysis of organic pollutants in the leachate is necessary, but it is very difficult because of the extremely complex nature of landfill leachate. An alternative way is to analyze the molecular weight distribution of organic compounds in the leachate by the GPC method.

In this study, an MBR with an air-lift bioreactor and gravity flow was applied to treating landfill leachate to investigate its performance of BOD$_5$, SCOD, and nitrogen removals. Another purpose of this study was to further understand the removal pathway of COD throughout the operational period of MBR treating landfill leachate by investigating changes of the molecular weight distribution of organic pollutants using the GPC method.

## 1   Materials and methods

### 1.1   MBR with air-lift bioreactor and gravity flow

The MBR was composed of a bioreactor with 80 L working volume and two membrane modules (Fig. 1). The membrane module was made of 0.22 μm hollow fiber of polyvinylidene fluoride (PVDF). Each membrane module with the area of 0.1 m$^2$ was placed outside the bioreactor, and connected to the bioreactor by two pipes with valves. Air was supplied from the bottom of the modules by an air pump. Aeration in this MBR had three functions: transferring oxygen to microorganisms, mixing the liquor and cleaning the membrane. The influent was fed into the bioreactor by a peristaltic pump. The permeation was driven by 9.0 kPa of the hydraulic pressure head between the level of mixed liquor in the bioreactor and the permeation outlet. The mixed liquor was carried by air into the central shaft-tube of the bioreactor, and then it rose up in the tube and came down outside the tube. So the influent was mixed and diluted by the recycle of the mixed liq-

uor. Due to the membrane modules connected to the bioreactor by pipes with valves, no direct discharge of the mixed liquor from the bioreactor is needed in the maintenance of the MBR, i.e. the MBR running was stopped by shutting the valves, and the membrane modules were disconnected from the bioreactor during cleaning or replacing membrane modules. As described above, this kind of MBR, with an air-lift bioreactor and gravity flow, has advantages of easy cleaning and maintaining of membrane modules, and energy saving[25]. Such MBR has been successfully applied in treating the municipal wastewater and dyeing wastewater[25,26].

### 1.2   Landfill leachates

Six landfill leachate samples were taken in December, 2002, and April to July of 2003 from A'suwei Municipal Landfill in the north of Beijing, China. This landfill site was started in 1996 and was still in use during the time of this study. After being taken from the landfill, the leachates were then stored in a storage tank at room temperature before it was pumped into the MBR. The characteristics of some typical leachates (Leachate I, taken in December, 2002; Leachate II, taken in May, 2003; and Leachate III, taken in July, 2003, respectively) listed in Table 2 indicated that the characteristics of landfill leachates varied with seasons. The concentrations of the contaminants (e.g. COD and NH$_4^+$-N) of the leachates taken in the spring and summer were much higher than those in the winter. Notably, the characteristics of landfill leachates changed during the storage time because of the microorganisms in the leachates. During the storage period, the variations of BOD$_5$ and SCOD in the leachates taken in the spring and summer were more than those taken in the winter because of the high BOD$_5$/SCOD ratio of the spring and summer leachates, but NH$_4^+$-N and TN (total nitrogen) did not change as much as BOD$_5$ and SCOD in all the samples (Table 3).
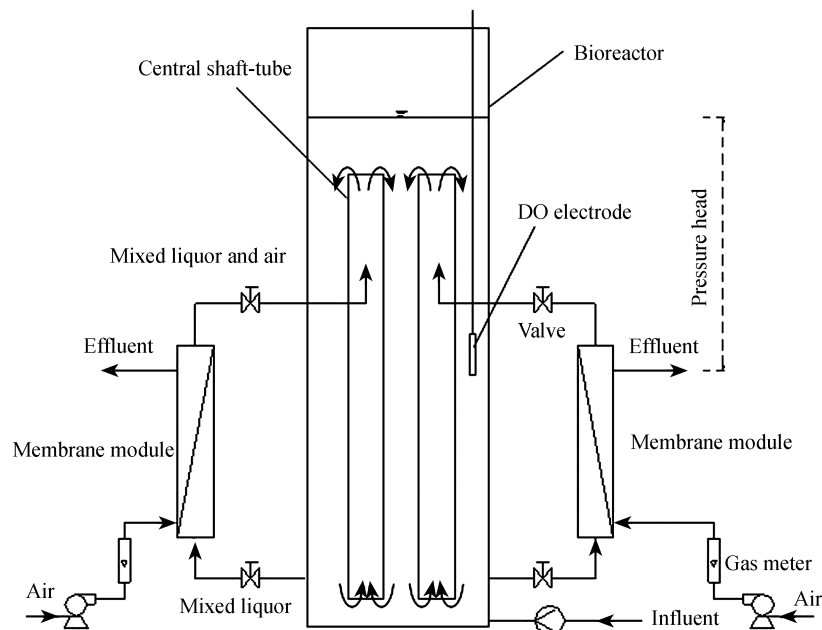
Fig. 1. Schematic diagram of the MBR with an air-lift bioreactor and gravity flow for landfill leachate treatment.

Table 2  Characteristics of landfill leachates I, II and III

|  | Leachate I | Leachate II | Leachate III |
| --- | --- | --- | --- |
| SCOD (mg/L) | 2049.6 | 11397.8 | 15526.3 |
| $BOD_5$ (mg/L) | 550 | 7200 | 9080 |
| $NH_4^+$-N (mg/L) | 1177.9 | 2346.0 | 1745.0 |
| $NO_3^-$-N (mg/L) | 14.6 | 32.6 | 39.1 |
| $NO_2^-$-N (mg/L) | 0.2 | 0.1 | 0 |
| TN (mg/L) | 1293.2 | 2445.0 | 1802.5 |
| pH | 8.5 | 8.0 | 8.5 |
| Conductivity (μs/cm) | 13000 | 25660 | 26400 |
| Total dissolved solids (mg/L) | 8454 | 16145 | 16280 |

Table 3  Variation of characteristics of landfill leachates during storage

|  | Storage time (d) | SCOD (mg/L) | $BOD_5$ (mg/L) | $NH_4^+$-N (mg/L) | TN (mg/L) |
| --- | --- | --- | --- | --- | --- |
| Leachate I | 22 | −337 | −100 | +68 | −19 |
| Leachate II | 7 | −3560 | −1800 | −52 | −19 |
| Leachate III | 14 | −5201 | −880 | −78 | −115 |

+, Increase; −, decrease.

### 1.3  Long-term running test

Some activated sludge taken from a municipal wastewater treatment plant was inoculated in this MBR. Within 60 d of the MBR start-up, the MBR was sequentially fed with the wastewaters combining with Leachate I and domestic sewage in the ratio of 4/1, 3/1, 2/1, 1/2, 1/3 (V/V), and Leachate I. The sharp increase of nitrification efficiency (from 13% on D 55 to 48.4% on D 60) meant the success of the MBR start-up, and then the long-term running test started. The experiment was carried out at ambient temperature (14−19℃ in the start-up period and 19−27℃ in the long-term running period). The two membrane modules were operated as follows: One was in the continuous running except for cleaning by air sparging (aeration intensity 600 $m^3/m^2$·h) for 12 h every 20 d. Its permeate flux was kept in the range of 6.7−9.5 $L/m^2$·h in the first 75 d. The other module was used as an accessory in order to regulate the hydraulic loading rate. The hydraulic retention time varied from 1.8 to 6.0 d on the basis of the variation of permeation flux before D 75. After D 75, HRT was controlled at the range of 6.0−12.9 d because of high COD of the leachate.

### 1.4  Molecular weight fractionation

The procedure of determining molecular weight fractionation of organic components in the landfill leachate was similar to the procedures proposed by Leidner et al.[27] and Millot et al.[28]. A chromatographic column (2.6×100 cm) was packed with the pre-swelled Sephadex gel G-50 (medium) (Amersham, Sweden). The column was calibrated by seven polyethylene glycols (PEG) (Merck, Germany) and $K_2CrO_4$ (Beijing Chemical Reagents Co., China) with the molecular weight (MW) of 20000, 10000, 3000, 1000, 400, 200, and 194.2 Da, respectively. The linear equation $\log(MW)=5.26-0.006V_e$ ($R^2=0.98$) was obtained, where MW was the molecular weight (Da); $V_e$ was the

elution volume (mL). The samples were filtered through a 0.45-μm membrane before passing the GPC column. The successive isomerous fractions of 10 mL filtrate were collected at the outlet of the column by a fraction collector (BSZ-100, Shanghai Qingpuhuxi Instrument Co., China). The DOC (dissolved organic carbon) concentrations of these fractions were determined by a TOC analyzer (Phoenix 8000 UV-persulfate TOC analyzer, Tekmar Dohrmann, USA). The U.V. absorbance at 254 nm (UV254) was measured by a UV/Vis spectrophotometer (Model 752, Shanghai Precision & Scientific Instrument Co., Ltd, China). Phosphate buffer solution (pH=8) at a flow rate of 30 mL/h was used as the eluent.

### 1.5 Analytic procedures

SCOD was determined for filtered samples (0.45 μm filters) using a COD analyzer (CTL-12, Chengde Huatong Instrument Co., China). $NH_4^+$-N, $NO_3^-$-N, $NO_2^-$-N, as well as TN were determined for filtered samples using a UV/Vis spectrophotometer (Model 752, Shanghai Precision & Scinetific Instrument Co., Ltd, China). TN analysis was done after digestion with a digester (VARIOKLAV steam sterilizer, H+P Labortechnik, Germany). $BOD_5$ was measured by a BOD analyzer (OxiTop, WTW, Germany). Dissolved oxygen (DO) in the bioreactor was measured by a DO meter (Oxi 330i, WTW, Germany).

## 2 Results and discussion

### 2.1 Removal of $BOD_5$ and COD

High $BOD_5$ removal efficiency of more than 99% was obtained during the MBR operation. At the loading rate of 2.43 and 1.71 $kgBOD_5/m^3 \cdot d$, $BOD_5$ in the effluent was less than 60 and 35 mg/L, respectively. $BOD_5$ of the supernatant of the mixed liquor in the bioreactor was slightly higher than that of effluent (Fig. 2(a)). More than 99% of $BOD_5$ removal efficiency indicated that there was still potential to increase $BOD_5$ loading rate of the MBR.

In spite of high $BOD_5$ removal efficiency in the MBR, COD removal was not as satisfactory as that of $BOD_5$ removal. Fig. 2(b) shows that the high SCOD (550 — 1790 mg/L) presented in the effluent. The SCOD removal efficiency varied between 72.3% and 96.2% correspondingly with changes of influent SCOD, but the impact of the SCOD loading rate on SCOD removal rate was not obvious. Results indicated that 5%—65% of supernatant SCOD in the bioreactor was removed by the membrane cut-off. Therefore, both SCOD of the supernatant in the bioreactor and the effluent were comparatively stable, although SCOD in the influent varied dramatically (4200—15900 mg/L) due to the landfill leachate taken in different seasons. These results showed that the COD concentration in the effluent was correlative to the character and molecular weight distribution of organic matters in the landfill leachate.

### 2.2 Removal of nitrogen

Biological removal of ammonium is one of the major objectives of the landfill leachate treatment because of high ammonium concentration in the leachate (Table 2). A dissolved oxygen (DO) difference was observed along the axial outside the central shaft-tube of the air-lift bioreactor, in which DO in the upper zone was
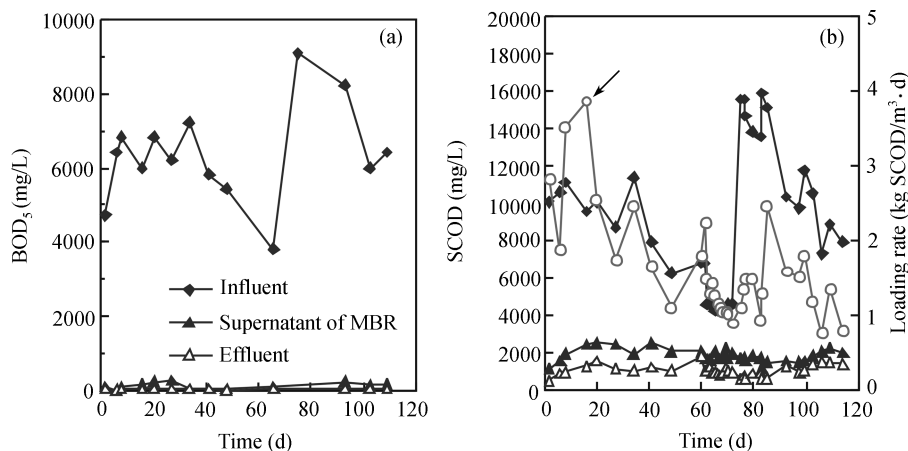


Fig. 2.  $BOD_5$ and SCOD removal in the MBR treating landfill leachate. (a) $BOD_5$ concentration; (b) SCOD concentration and loading rate.

higher than that in the lower zone, especially during MBR running at high $BOD_5$ loading rate and low aeration intensity. Hence it was possible for the nitrification and denitrification to take place simultaneously in the bioreactor, but it is very difficult to realize the stable simultaneous nitrification and denitrification due to sharp fluctuation of the influent concentration.

Throughout the long-term running test, three peaks of ammonium concentration in the effluent were observed due to the following three different factors (Fig. 3(a)). Firstly, the $NH_4^+$-N loading rate abruptly increased to 0.95 kg $NH_4^+$-N/$m^3$·d on D 16 from 0.37 kg $NH_4^+$-N/$m^3$·d on D 6, but the aeration intensity was not increased correspondingly in time, and then resulted in less than 0.5 mg/L of the DO in the middle zone outside the central shaft-tube of the bioreactor (The position of the DO sensor shown in Fig. 1. DO values shown in Fig. 3(b)). Hence the nitrification was severely inhibited. Secondly, the $NH_4^+$-N loading rate increased drastically from 0.40 to 0.81 kg $NH_4^+$-N/$m^3$·d from D 48 to D 64, while the DO in the middle zone of the bioreactor was at the range of 1.0 to 2.0 mg/L. In this case, the MBR faced with overload of $NH_4^+$-N and the nitrification was suppressed again. Therefore, the high concentration of $NH_4^+$-N (more than 1000 mg/L) occurred in the effluent and only 20%－30% of TN was removed. In addition, control of DO to reach simultaneous nitrification and denitrification in D 85－102 may be the reason of the third peak occurrence (129－704 mg/L of $NH_4^+$-N in the effluent). TN in the effluent was 611－750 mg/L and its removal efficiency was 55.5%－70.8% at the DO of 0.5－1.5 mg/L and $NH_4^+$-N loading

rate of 0.17－0.28 kg $NH_4^+$-N/$m^3$·d, respectively. After D 106, the $NH_4^+$-N in the effluent was below 15 mg/L and the nitrification product was almost nitrate, when DO and $NH_4^+$-N loading rate were controlled at the range of 2.3－2.8 mg/L and 0.16－0.24 kg $NH_4^+$-N/$m^3$·d, respectively. On the other hand, the denitrification rate decreased due to high DO in the MBR, and as a result, the TN removal efficiency decreased from 55.5% on D 106 to 44.5% on D 114 (Fig. 3(b)).

It is well known that oxygen is one of the key factors of nitrification and denitrification. If DO concentration is low, the ammonium can only be oxidized to nitrite or the nitrification process will even stop. Otherwise, the denitrification could be inhibited when DO is high. Another key factor is $NH_4^+$-N loading rate. At less than 0.24 kg $NH_4^+$-N/$m^3$·d, two events took place in this study. One was that simultaneous nitrification and denitrification existed significantly at about 1 mg/L of DO, the other was that the $NH_4^+$-N was oxidized to nitrate completely at over 2 mg/L of DO.

### 2.3 Molecular weight fractionation

As discussed above, the removal efficiencies of $BOD_5$ and $NH_4^+$-N were excellent when the leachate was treated in the MBR under optimal conditions. However, SCOD in the effluent was still high despite the membrane filtration. In order to study this phenomenon, the changes of organic matter molecular weight during MBR treating landfill leachate were investigated by means of GPC, and Leachate I, Leachate II (fed into the MBR from D 34 to D 61) and Leachate III (fed into the MBR from D 75 to D 98) were studied in this investigation. The GPC profiles of leachate represented a
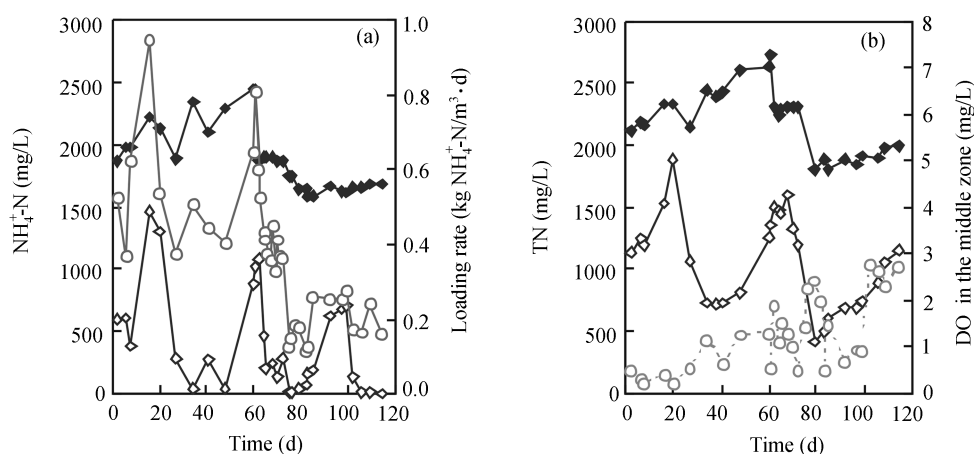


Fig. 3.　Removal of $NH_4^+$-N and TN in the MBR treating landfill leachate. (a) $NH_4^+$-N concentration and loading rate. ◆, $NH_4^+$-Nin;　◇, $NH_4^+$-Nout; ○, loading rate; (b) TN concentration and DO in the middle zone of the bioreactor.　◆, TNin;　◇, TNout; ○, DO.
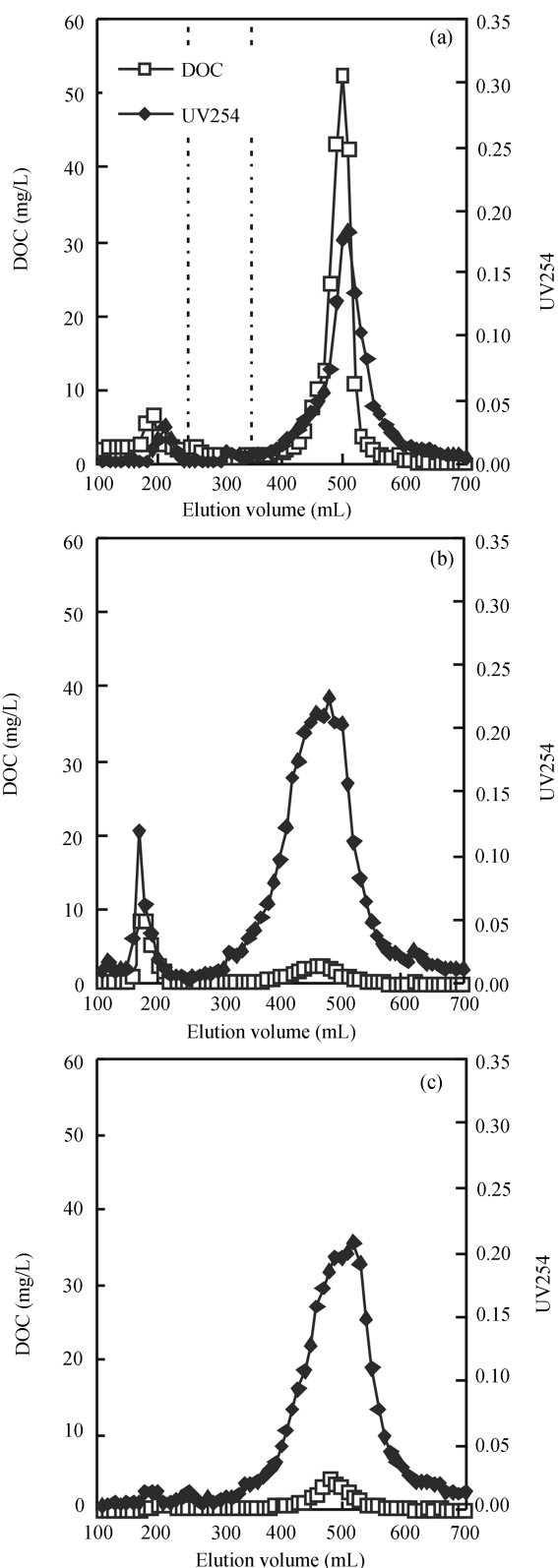
Fig. 4. GPC profiles of Leachate II throughout the MBR treatment characterized by DOC and UV254. (a) Raw leachate; (b) supernatant of the MBR; and (c) effluent.

bimodal distribution characterized by either DOC or UV254 (Fig. 4(a)), which was similar to the results of Chain and DeWalle[29] and Harmsen[30]. These landfill leachates can be divided into three molecular weight fractions by GPC as follows: 1) Fraction A, $V_e$=100—250 mL, $MW$ >5754 Da, $MW$p = 11480—13182 Da; 2) Fraction B, $V_e$ = 260—340 mL, $MW$ = 1445—5754 Da; and 3) Fraction C, $V_e$ = 350—700 mL, $MW$<1445 Da, $MW$p = 158—275 Da (Fig. 4(a)). As shown in Fig. 5(a), the leachates mainly consisted of two kinds of organic matters on the basis of the molecular weight distribution, namely high molecular weight fraction (Fraction A) and low molecular weight fraction (Fraction C). Organic matters of Fraction C contributed much more to DOC concentrations than those of Fraction A and B in the raw landfill leachates.

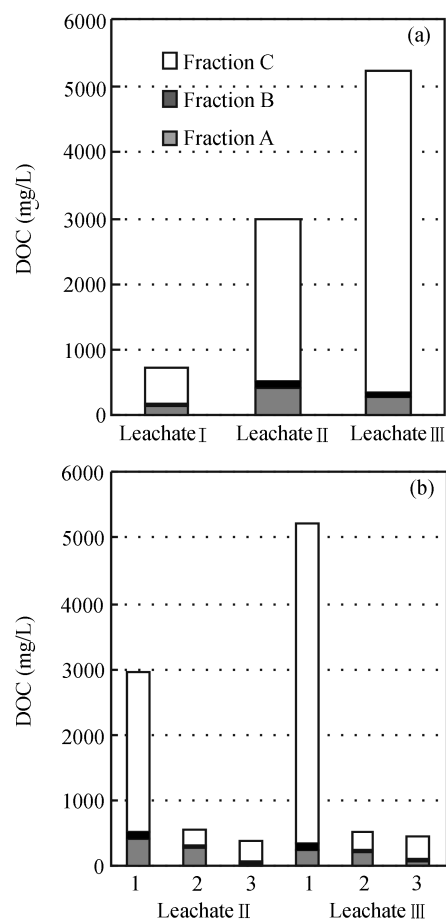As shown in Fig. 5(a), DOC of Fraction A in different seasons was relatively stable (136.4, 432.3 and



Fig. 5. Molecular weight distribution of landfill leachate characterized by DOC. (a) Landfill leachate in different seasons; (b) landfill leachate throughout the MBR treatment. 1, Raw leachate; 2, supernatant of the MBR; 3, effluent.

266.6 mg/L of Leachates I, II and III, respectively), while DOC of Fraction C in different seasons fluctuated dramatically (554.7, 2400.8 and 4892.7 mg/L of Leachate I, II and III, respectively). The proportion of Fraction A in Leachate I (18.9%) was higher than that in other two samples (14.7% and 5.1% of Fraction A in Leachate II and III, respectively), and Leachate III had the highest percentage of Fraction C (76.9%, 81.5% and 93.4% of Fraction C in Leachates I, II and III, respectively).

All the organic matters of the three fractions decreased after biological treatment, but the amounts of reduction were different. As shown in Fig. 5(b), most of DOC in Fraction C was removed from 2400.8 to 254.3 mg/L of Leachate II, and from 4892.7 to 289.3 mg/L of Leachate III. At the same time, DOC in Fraction A was recalcitrant to be biodegraded (from 432.3 to 291.9 mg/L of Leachate II, and from 266.6 to 221.9 mg/L of Leachate III) and resulted in increasing of the proportion of Fraction A in the supernatant of the bioreactor. As it was more effective for the membrane to cut off the organic matters in Fraction A than those in Fraction C, the proportion of Fraction A in effluent decreased obviously after membrane filtration.

Fig. 4 shows the GPC profiles of Leachate II detected by a DOC analyzer and a UV spectrophotometer at 254 nm (the characteristic absorption of aromatic cyclic compounds), respectively. UV254 absorbance curve in the raw leachate changed accordingly with the DOC curve (Fig. 4(a)), but their trends were very different after biological treatment and membrane cut-off. After aerobic biological treatment, the DOC of the Fraction C was decreased dramatically, while UV254 absorbencies of the Fractions A and C were increased slightly (Fig. 4(b)). These results implied that the aerobic biological treatment was inefficient for removing aromatic cyclic compounds. Most of the aromatic cyclic compounds in the Fraction A were removed as a result of the membrane cut-off, but those in the Fraction C passed through the membrane and then presented in the effluent (Fig. 4(c)).

Wichitsathian et al.[23] reported that low molecular weight compounds are composed of easily degradable volatile fatty acids and amino acids. Medium molecular weight compounds having a molecular weight between 500 and 10000 Da contain fulvic acid-like substances and compounds with carboxylic and aromatic hydroxyl groups. High molecular weight compounds consist of carbohydrates, proteins, and humic-like substances.

Fraction A may be mostly composed of carbohydrates, proteins and humic-like substances. These high molecular weight compounds are refractory for biodegradation, but most of them can be cut off by membrane. Fraction C may be composed of volatile fatty acids, amino acids, fulvic acids and compounds with carboxylic and aromatic hydroxyl groups. Volatile fatty acids and amino acids are easily biodegradable, so DOC of Fraction C is decreased after aerobic treatment. The residue in Fraction C may be fulvic acid and compounds with carboxylic and aromatic hydroxyl groups. These organic compounds not only are difficult to be biodegraded, but also can pass through the membrane, thus causing high SCOD in the effluent.

## 3 Conclusions

High removals of $BOD_5$ and $NH_4^+$-N were achieved in an MBR with an air-lift bioreactor and gravity flow treating landfill leachate under optimized conditions. However, the removal efficiency of SCOD was not as high as that for $BOD_5$ removal rate.

The investigation of organic matter molecular weight distribution by GPC indicated that organic matters of the raw landfill leachate were composed of a high MW fraction and a low MW fraction, and the low MW fraction contributed more to DOC than the high MW fraction. The high MW fraction was recalcitrant to be biodegraded, but could be removed by the membrane cut-off. Though most of the low MW fraction was biodegradable, the refractory low MW fraction was able to pass through the membrane, thus resulting in high SCOD in the effluent.

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