Behavior of dibutyl phthalate in a simulated landfill bioreactor

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ABSTRACT

In this study, the behavior of dibutyl phthalate (DBP) from municipal solid waste (MSW) in the leachate and refuse of two simulated landfill bioreactors was compared. In one reactor, the leachate was circulated between a landfill and a methanogenic reactor, while the other reactor was operated using direct recirculation of the leachate. The results revealed that the original concentration of DBP in the refuse was approximately 18.5 μg/g, and that this concentration decreased greatly during decomposition of the waste for both reactors. Furthermore, the major loss of DBP from the landfill occurred in an active methanogenic environment in the later period, while the environment was acidic due to a high concentration of volatile fatty acids (VFA) and contained a large volume of biologically degradable material (BDM) during the early stage. Circulating the leachate between the landfill and a methanogenic reactor resulted in an increase in the biodegradability of MSW and a high degree of waste stabilization. Furthermore, DBP degraded more rapidly in the landfill that was operated in conjunction with the methanogenic reactor when compared to the landfill in which there was direct leachate recirculation.

1. Introduction

Although phthalic acid diesters (PAEs) are used in a wide variety of products, their primary use is in polyvinylchloride (PVC) as plasticizers [1]. PAEs easily bioaccumulated in the apuatic and the terrestrial environment and some PAEs are considered to be potential carcinogens, teratogens and mutagens [2]. Among PAEs, dibutyl phthalate (DBP) is one of the most widely used, and its consumption is growing rapidly both globally and locally in China [3–5]. DBP is also the most frequently identified PAE in environmental samples, and it has been listed as a priority pollutant by the US EPA and its counterparts in China, Japan and some European countries [6–8].

Currently, most materials containing DBP are disposed of with other municipal solid waste (MSW) by landfill. Landfills pass through typical phases soon after waste is deposited [9]. First, the landfill becomes anaerobic due to the depletion of oxygen, during which organic compounds are hydrolysed and fermented to primarily volatile fatty acids (VFA). In the second phase, methanogens begin to proliferate, and acetogenic bacteria convert the VFA to acetic acid, hydrogen and carbon dioxide. In the final and longest phase, these biodegradation intermediates serve as substrates for the production of methane via methanogens. Throughout this process, organic materials are released by degradation of the waste, which gives rise to a high level of organic matter in the leachate. DBP

is only physically bound to the plastic structure, therefore, it may be released from its products and easily leach out to the environment.

DBP is relatively stable in the natural environment. It has been shown that hydrolysis, photolysis and volatilization of DBP occurred very slowly, while biodegradation plays an important role in the decomposition of DBP [10–12]. Indeed, studies of DBP in seawater, soil, activated sludge and a constructed wetland revealed that it was degraded most rapidly in response to biodegradation [13–17]. However, degradation of DBP in a landfill is more difficult due to the complex anaerobic environment. Jonsson et al. [18] investigated the degradation of DBP to monobutyl phthalate and phthalic acid only under methanogenic conditions in a landfill. Besides, several laboratory studies have been conducted to evaluate transformation dynamics of DBP and its metabolites [19,20]. The results of these studies suggested that the degradation phase of a landfill determines the extent of PAEs metabolism. Additionally, Mersiowsky et al. [21] confirmed that biodegradation plays an important role in the fate of DBP under landfill conditions. However, these studies have focused on transformation of DBP in leachate from simulated landfill reactors that were operated under conventional sanitary landfill conditions.

Sanitary landfills that employ operational techniques such as leachate recirculation represent an economical and environmentally acceptable method for the disposal of MSW [22]. Leachate recirculation is capable of permanently reserving additional carbon in the landfill, which results in higher levels of methane generation as well as leachate treatment in situ. However, if the MSW contains a high proportion of easily digestible materials, the increased level

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of biodegradation associated with leachate recirculation can result in an imbalance in the growth rates of rapidly growing acidogenic bacteria and slow growing methanogens during the first phase of MSW decomposition. Such an imbalance can result in methanogenesis being delayed or inhibited [23–25]. Therefore, many studies have been conducted to evaluate the use of treated leachate recirculation to accelerate refuse decomposition [26–28]. The results of these studies have indicated that bioreactor landfills (BLs) that circulate the leachate between the landfill and a methanogenic reactor take advantage of adapted microflora and the highly alkaline effluent of the methanogenic reactor to buffer the pH and inoculate the landfill. The addition of this effluent to the landfill can create optimal environmental and nutrient conditions for acidogenic bacteria and methanogens, thereby improving the overall performance of the system. However, to date, there have been no studies conducted to evaluate the behavior of DBP in these types of landfills.

This study was conducted from the viewpoint that bioreactor landfills are superior to conventional landfills and they can provide an advantage for the transformation of organic materials. Accordingly, we evaluated the behavior of DBP in simulated landfill bioreactors. To accomplish this, we analysed the occurrence and degradation of DBP in leachate from simulated landfill bioreactors. In addition, we analysed the pH, chemical oxygen demand (COD) and VFA of the leachate and biologically degradable material (BDM) of MSW to achieve a general characterization of the landfill. Finally, we compared the behavior of DBP in landfill bioreactors operated under different modes.

2. Materials and methods

2.1. Experimental set-up

Diagrams of the simulated landfill bioreactors used in this study are shown in Fig. 1. The BL was comprised of a methanogenic reactor that received leachate from the landfill. In this system, the leachate was subjected to methanogenesis in the methanogenic reactor, after which it was recycled into the landfill. Leachate was continuously circulated within 8 h daily between the landfill and the methanogenic reactor by using pumps with adjusted flow rates varying with leachate volume during waste decomposition, except for the first 2 days when no recycled lechate was fed to the landfill reactor. The recycling ratio of leachate was 100%, therefore the recycled leachate volume, i.e. the effluent volume, nearly amounted to the influent leachate volume every day. A leachate direct recirculation landfill (RL) was used as a control. To avoid uncontrolled contamination with DBP, only tubing and pump components made of PTEE were used.

Both landfill bioreactors were constructed of brick-concrete and had the same effective size of 0.55 m × 0.55 m × 2.0 m ($L \times W \times H$). A plexiglass male adapter was installed at the bottom of each landfill bioreactor as a leachate drainage port. In addition, two such adapters were installed in the lid of each landfill bioreactor to enable leachate recirculation and gas collection. Furthermore, two MSW sampling ports were installed on one side of the landfill. The methanogenic reactor, which was constructed of plexiglass and had a working volume of 15 l, was seeded with 12 l of raw anaerobic sludge obtained from the Hangzhou Sibao sewage treatment plant. The sludge was incubated with the synthetic wastewater with a COD of 3000 mg/l for 10 days to activate the sludge activity. The synthetic wastewater contained (g/l): saccharose, 2.75; NH_4Cl, 0.03; KH_2PO_4, 0.73; $KHPO_4$, 0.25; $NaHCO_3$, 3.30. Then it was acclimated by the leachate with COD concentration of 2625 mg/l from Hangzhou Tianziling landfill site. The start-up of methanogenic reactor was accomplished when the COD removal efficiency reached stability at above 80% under organic loading rate of about 2.5 kg COD/m^3 day. The acclimated sludge in the

methanogenic reactor had a total solids (TS) content of 90.5 g/l and a volatile solids (VS) content of 27.0 g/l.

Fresh refuse was collected from the Kaixuan transport station in Hangzhou, Zhejiang, east China. The physical composition of the refuse (by weight) was as follows: kitchen waste, 61.5%; plastics, 11.6%; paper, 10.3%; sand and soil, 7.1%; cellulose textile, 1.3%; glasses, 6.4%; metals, 0.6%; rubber, 0.6%; wood, 0.6%. The moisture content of the refuse was about 54%. Prior to adding the refuse to the landfill bioreactor, a 5 cm thick layer of gravel was placed at the bottom of the reactor to retain refuse and prevent small particles from leaching out. In addition, larger particles of the collected refuse were shredded into 2 cm approximately and thoroughly mixed. Then about 200 kg refuse, which was added with tap water to 75% moisture content, was filled into landfill reactor. Therefore the wet density of the refuse compacted in the landfill bioreactors was 600 kg/m^3. After the water was added, the MSW was covered with a 5 cm layer of sand so the leachate would be well distributed when it was recycled. Finally, the bioreactors were sealed airtight.

2.2. Sampling procedure

Leachate samples were collected from the landfill leachate drainage port and methanogenic reactor outlet (Fig. 1) daily to determine the pH. In addition, the COD and VFA of the leachate samples were determined weekly. Furthermore, BDM of refuse samples collected periodically from the landfill was also determined. Both simulated landfill reactors were operated at room temperature and the methanogenic reactor were carried out in a temperature-controlled room at 30 ± 1 °C for 310 days.

To evaluate changes in the DBP in the landfill bioreactors, leachate samples were collected from the sampling point using a glass tank. The leachate samples were then immediately transferred to brown glass bottles and analysed for DBP. Refuse samples were withdrawn from the upper and lower layer of each landfill reactor. After mixing by hand manipulation, refuse samples were further cut and ground in several steps to 20 meshes. Finally they were freeze dried and stored at −20 °C until analysis. To avoid contamination of the samples by laboratory equipment during handling and analysis, all glassware was washed and rinsed in ethanol and subsequently heated at 450 °C for 20 h [29]. In addition, blank levels of DBP were monitored during each series of sample preparation and considered in the corresponding calculations. All analyses of DBP in the leachate and refuse were conducted in triplicate to ensure the validity of the results.

2.3. Analytical methods

Degradation of DBP was assessed by measuring the disappearance of the parent chemicals by high performance liquid chromatography (HPLC) using the methods described in the standard methods ("Water quality–Determination of phthalate (dibutyl)–Liquid chromatography", State Environmental Protection Administration of PR China, 2001), with some modification. Briefly, 50 ml samples of leachate were extracted three times with hexane (2:1, v/v), after which the pH value of the sample was adjusted to 7–8 with 1 N NaOH or 1 N HCl if necessary. The hexane extracts were then passed through a small glass hopper containing Na_2SO_4 to eliminate contaminating water. Furthermore, the method of Li [30] was used for pretreatment and extraction of the DBP from the refuse with some modification. Briefly, 25 g sample of refuse (20 meshes) was placed into a conical flask of 500 ml capacity, and 250 ml of redistilled water was added. The flask was shaken vigorously on a mechanical shaker for 24 h (200 rpm). After standing for 48 h, liquor phase was filtrated with a membrane (0.45 μm). The procedure of extraction with hexane like above was repeated three
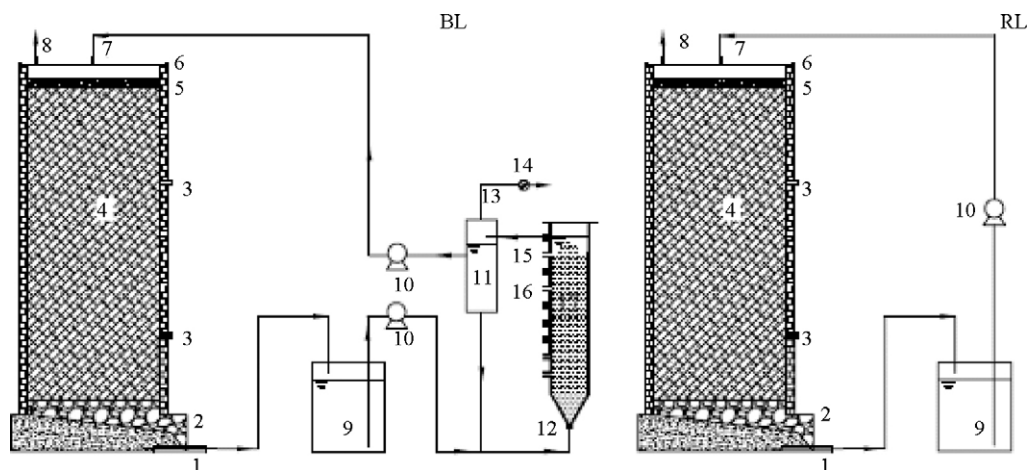
Fig. 1. Schematic diagram of the landfill bioreactor system. 1. leachate outlet, 2. gravel layer, 3. MSW sampling port, 4. landfill site, 5. sandy layer, 6. headspace, 7. vent-port, 8. gas outlet, 9. leachate collection tank, 10. peristaltic pump, 11. seperation gas from liquid, 12. leachate inlet, 13. gas outlet, 14. wet gas meter, 15. outlet, 16. sludge sampling port, 17. methanogenic reactor.

times. Next, the extracts were concentrated to 1 ml using a K–D concentrator prior to analysis using an HPLC (Agilent 1100, USA). This procedure was performed in triplicate. The samples were then injected through an AE LICHROM column (CN-5 μm) using a mixture of hexane and isopropanol mixture (99:1, v/v) at a flow rate of 1.5 ml/min as the mobile phase. DBP was then detected using a UV detector at a wavelength of 272 nm. The detection limit was 0.1 μg/l for DBP.

The COD and pH in the leachate were determined using the standard methods [31]. VFA was analysed using the acidified ethylene glycol colorimetric method [32]. The BDM of the MSW was analysed using the potassium dichromate method [33].

3. Results and discussion

3.1. Variation of DBP concentrations in leachate and refuse

Fig. 2 shows changes in the concentrations of DBP in the leachate over time. The initial DBP concentrations in the leachate were much higher than the concentrations reported in previous studies (Table 1). This difference may have occurred due to differences in the age and components of the waste in the source landfills used in each study. In this study, the concentrations of DBP ranged from approximately 1100–2100 μg/l during the early period. This fluctuation may indicate that the DBP underwent slight degradation during the early stage, but the leaching of it varied its concentrations in leachate again. This result is similar to the findings of a study that was previously conducted by Bauer and Herrmann
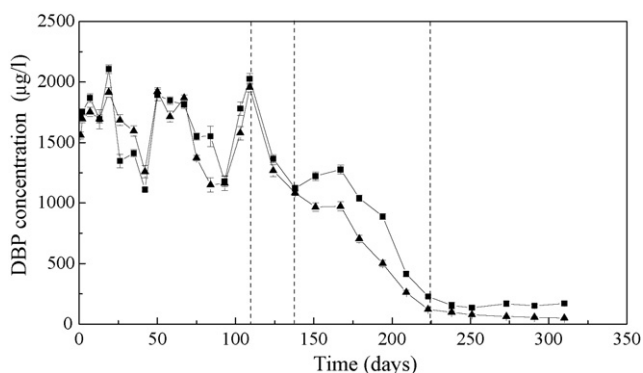
[36]. However, an apparent decrease in the DBP concentrations was observed after day 110. This decrease in concentration indicates that environment of the landfill became more suitable for DBP degradation during the later period. In addition, DBP was degraded more rapidly in the BL than in the RL during that period. This difference was likely due to the different conditions in the two reactors. However, the DBP was not degraded exhaustively, and was still present at concentrations of approximately 150 and 60 μg/l in the RL and BL, respectively, at the end of experiment. This may have occurred due to the continuous leaching behavior of DBP. This finding is similar to those of previous studies that found DBP was still present in the leachate from landfills several years after closure [37].

Refuse samples were collected from the upper and lower layer of the landfill to evaluate variations in the DBP concentrations. As shown in Fig. 3, the background content of DBP in the refuse was approximately 18.5 μg/g, and this value decreased greatly during waste decomposition, especially from day 50 to day 200. In addition, as in the leachate, DBP degradation occurred more rapidly in the BL than in the RL. These differences may have occurred due to different landfill conditions. DBP degradation coincided with the general degradation of organic material, which likely changed the particulate and colloidal properties of the organic matter, thereby changing the sorption characteristics, and influencing the DBP transformation [38]. Similarly, the residual DBP in the MSW would lead to the continuous leaching behavior.

3.2. Effects of landfill stabilization process on the behavior of DBP

As shown in Fig. 2, the major loss of DBP from the landfill bioreactors occurred during the later period, and the degradation of DBP

Fig. 2. Variation of DBP concentrations in leachate from RL (■) and BL (▲).

Table 1
DBP concentrations in leachate of previous studies and this experiment.

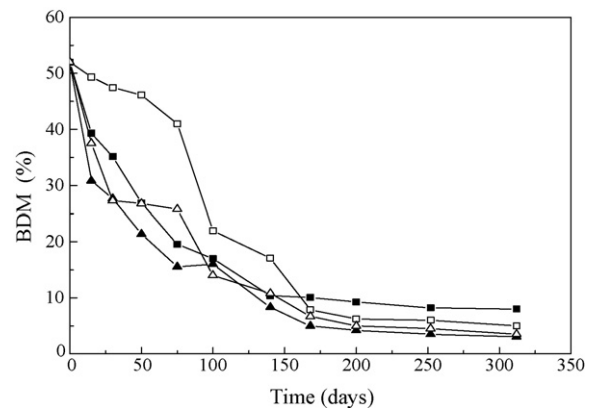| Maximum concentration of DBP (μg/l)	Source of leachate	Waste type	References
23	Landfill sites	MSW, industrial waste	[18]
255	Simulated landfill reactors	MSW	[20]
15	Landfill sites	MSW	[34]
1400	Landfill sites	MSW	[35]
2106	Simulated landfill reactors	MSW	In this study
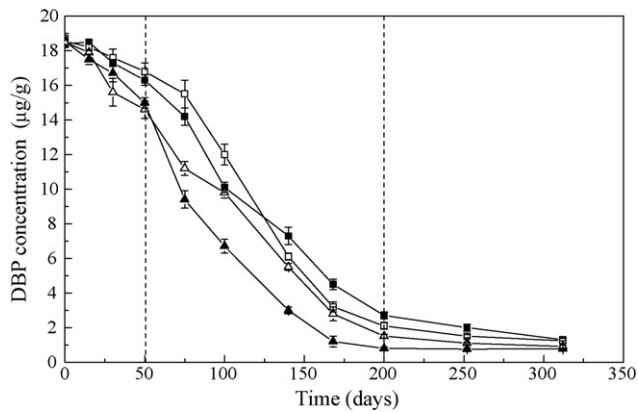
Fig. 3. Variation of DBP concentrations in refuse from RL and BL. RL upper (■); RL lower (□); BL upper (▲); BL lower (△).

Fig. 5. Variation of BDM of refuse from the upper and lower layers of RL and BL. RL upper (■); RL lower (□); BL upper (▲); BL lower (△).

was faster in the BL than in the RL. These findings may indicate that the landfills had different stabilization processes.

The leachate characteristics are known to mirror the biodegradation of the organic refuse and the process of landfill stabilization [39]. Changes in the pH, COD and VFA of the leachate from the two bioreactors over time are shown in Fig. 4. The pH of the leachate from the RL and BL increased from acidic to approximately neutral after 120 and 85 days, respectively. In addition, the COD concentrations of the leachate from the RL were higher than those from the BL during the early experimental period, and they stabilized at approximately 2500 mg/l after 150 and 120 days, respectively. These findings suggested that circulating the leachate between a landfill and a methanogenic reactor may accelerate waste stabilization. A similar phenomenon was observed when the VFA was

**Fig. 4.** Variation of pH, COD and VFA of leachate from RL and BL. COD (□); VFA (◇); pH (△).

evaluated. Specifically, the VFA concentrations of the leachate from the RL and BL increased to 27,720 mg/l and 16,940 mg/l after 75 days, respectively. These levels resulted in the pH of the leachate from the RL and BL being 5.9 and 6.8, respectively. After 140 day, the VFA concentration had decreased to less than 200 mg/l in the leachate from the BL, indicating that the BL was completely methanogenic at that time [40]. However, the concentration of VFA was still fluctuating between 200 and 450 mg/l in the RL at 140 days. Taken together, these results indicate that the pH values of the leachate from both bioreactors approached neutral and the COD concentrations were maintained at a low level for a long time during the latter period. These findings suggest that the two bioreactors had stabilized, but the environment in the BL was more stable than the environment in the RL.

Of the parameters evaluated in this study, the BDM best reflects the degree of biodegradation of the MSW [41]. Fig. 5 shows the variation in the BDM of the MSW from the upper and lower layers of both bioreactors. The degradation rate of MSW from the upper layer of the BL was higher than that of MSW from the upper layer of the RL. Indeed, by the time the COD concentrations of the leachate had risen to their highest value, the BDM value of the upper layer refuse had decreased from 52.0 to 19.6% and 15.6% in the RL and BL, respectively. This finding may indicate that the environment of the upper layer refuse was more suitable for degradation by the predominate microbes in the BL than by those in the RL. The BDM in the lower layer of refuse in the RL was much higher than that of the lower layer of refuse in the BL during the early period. This finding may be ascribed to re-adsorption of the organic substances by the lower layer refuse when the raw leachate was recycled in the RL. After 175 days, the BDM was maintained at a low level, which suggests that both of these bioreactors had entered into the stabilized phase. However, the BDM of refuse from the BL was lower than that of the RL during the later period, which indicates that the BL had a better degradation environment and stabilized earlier than the RL.

Fig. 6 shows the relationship between DBP concentrations and the BDM of refuse. The DBP residual concentrations and the BDM of the refuse were found to be highly correlated in both systems. Specifically, the correlation coefficients (R) were 0.9070, 0.9848, 0.9098 and 0.9397 for the upper layer of the RL, the lower layer of the RL, the upper layer of the BL and the lower layer of the BL ($P < 0.01$), respectively.

No obvious decrease in DBP was observed in the leachate during the early period. This finding indicates that, although DBP was released from plastics and was present in the acidic leachate, the microflora were not able to degrade it rapidly. However, a larger decrease in the DBP was observed under methanogenic conditions (Figs. 2 and 3). Similarly, previous degradation assays of the DBP
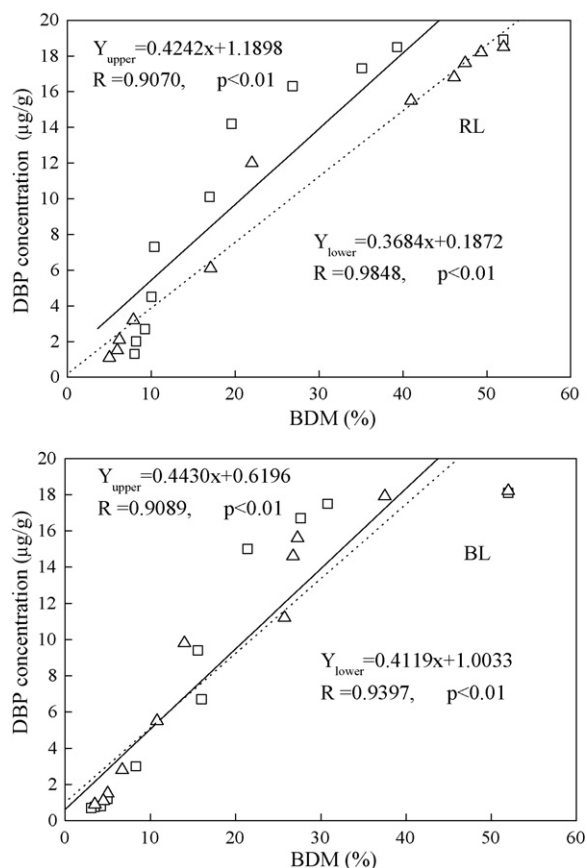
Fig. 6. Correlations of DBP concentrations and BDM of refuse from RL and BL. Upper (□, —); lower (△, ⋯). Note: DBP concentrations were the mean value.

in the reactors revealed that the potential for the acidogenic waste to degrade phthalate diesters was low. Taken together, these findings indicate that the degradation of DBP primarily occurred in the methanogenic waste [21,37,42].

3.3. Comparison of the behavior of DBP in the RL and the BL

As mentioned above, the DBP was degraded more rapidly in the BL than in the RL. This may have occurred due to the combined effects of the methanogenic reactor and the landfill reactor. Throughout the entire experiment, there were no detectable VFA in the effluent of the methanogenic reactor, and the pH values were all approximately neutral. Fig. 7 shows the COD concentrations and COD removal efficiencies in the methanogenic reactor. The COD removal efficiencies were maintained at great than 90% until day 95. However, the efficiency declined as the influent COD concentrations decreased, with the efficiency being approximately 50% at the end of the experiment. This likely occurred due to the low biodegradability of organic materials in the leachate from the old landfill [43].

A similar phenomenon was observed when DBP removal in the methanogenic reactor was evaluated (Fig. 8). The DBP removal efficiencies were maintained at approximately 90% during the early period, when the influent DBP concentrations were high. However, the efficiency then declined as the influent DBP concentrations decreased. Indeed, the DBP concentrations in the effluent of the methanogenic reactor decreased from 150 to 30 μg/l during the experiment. These findings indicate that the methanogenic reactor had great potential for degrading DBP, especially when the concentrations of DBP were relatively high.
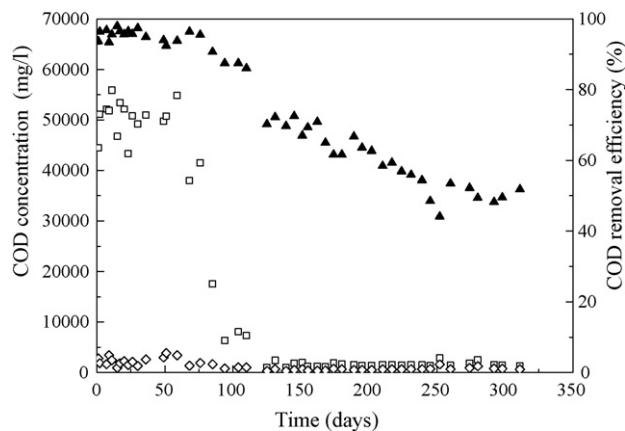
Fig. 7. COD concentrations and removal efficiencies in the methanogenic reactor. Influent (□); effluent (◊); removal efficiency (▲).

The results presented above demonstrate that the organic matter had already degraded to some extent when the leachate was fed into the methanogenic reactor. Once in the reactor, the methanogens produced enough alkalinity to buffer the acidic conditions caused by VFA in the landfill. Conversely, the refuse in the RL stabilized slowly due to the low pH value and high VFA concentration in the circulating leachate, which may have inhibited methanogenesis. Taken together, these findings indicate that the landfill surrounding was more suitable for the degradation of organic substances in the BL, which resulted in DBP being degraded more rapidly in the BL than the RL.

It was difficult to accurately calculate the degradation rate of DBP in the leachate or the refuse because the degradation and leaching behavior of DBP occurred simultaneously during refuse decomposition. The DBP concentrations of leachate dropped obviously from 1000 μg/l on day 140 to 60 μg/l till the end of the study, while they ranged from approximately 1100–2100 μg/l before that time in the BL (Fig. 2). As mentioned above, the BL was completely methanogenic after 140 days. This variation in concentration indicates that the loss of DBP primarily occurred in the methanogenic phase of the landfill. Furthermore, by paired samples test of DBP concentrations of leachate in two reactors, we found the DBP concentrations of leachate in the BL were significantly lower than those of leachate in the RL after 140 days ($P < 0.01$). These findings may indicate that DBP degradation occurred more rapidly in the BL than in the RL.
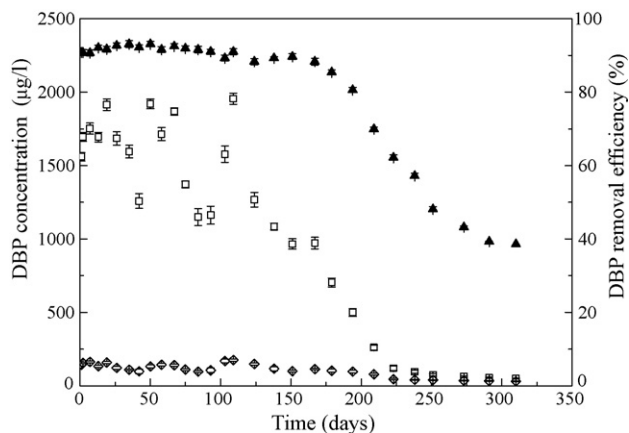
Fig. 8. DBP concentrations and removal efficiencies in the methanogenic reactor. Influent (□); effluent (◊); removal efficiency (▲).

Table 2
DBP concentrations and settlements of refuse for the RL and BL.

Time (days)	DBP concentrations of refuse (μg/g)				Settlement (%)	
	RL upper	RL lower	BL upper	BL lower	RL	BL
Initial ($t = 0$)	18.40 ± 0.22	18.55 ± 0.34	18.53 ± 0.28	18.48 ± 0.20		
Final ($t = 310$)	1.30 ± 0.07[a]	1.25 ± 0.17[a]	0.77 ± 0.05[b]	0.91 ± 0.08[b]	12	24

Parameters followed by different letters (a, b) are significantly different at $P < 0.05$ level.

In addition, the residual DBP concentrations of refuse in the BL were significantly lower than those of refuse in the RL. The DBP concentrations of refuse were approximately 0.7–0.9 μg/g in the BL at the end of experiment, while they were 1.2–1.3 μg/g in the RL at that time. Furthermore, the corresponding cumulative settlements in the RL and BL were 12 and 24% of the initial refuse height at the end of the study (Table 2). This indicates that circulating the leachate between a landfill and a methanogenic reactor resulted in an increase in DBP biodegradation and a high degree of waste stabilization.

4. Conclusions

The results of the analysis of DBP in the leachate and refuse from the simulated landfill bioreactors indicate that the landfill stabilization phases affected the behavior of DBP in the refuse. The loss of DBP from the landfill was much higher in an active methanogenic environment than in an acidic environment with a high VFA concentration and BDM. In addition, the results of this study revealed that circulating the leachate between a landfill and a methanogenic reactor resulted in an increase in the biodegradability of MSW and a high degree of waste stabilization. Finally, DBP was degraded more rapidly in the bioreactor landfill that was connected to a methanogenic reactor, and the removal of DBP was enhanced in the landfill with the methanogenic reactor than in the landfill with direct leachate recirculation.

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