Effects of leachate recirculation on anaerobic treatment of municipal solid waste

Chayanon Sawatdeenarunat

ABSTRACT

Anaerobic bioreactor landfills are the most popular method to treat municipal solid waste. Leachate recirculation is one of the methods to apply in bioreactor landfills to eliminate the leachate treatment and ground water contamination. The leachate recirculation could enhance landfill's performance both in term of municipal solid waste (MSW) treatment and landfill gas production. The waste degradation is accelerated by the leachate recirculation as well as the landfill's gas production rate is higher. Moreover, the proportion of methane in landfill gas is higher with leachate recirculation than those without recirculation. Nevertheless, chloride and ammonia nitrogen might accumulate in landfills and could inhibit methanogen bacteria. A key point of the recirculation rate of the bioreactor landfill is the balance of hydraulic pattern and biological process.

KEYWORDS

Municipal Solid Waste, bioreactor Landfills, Leachate recirculation, Anaerobic treatment

INTRODUCTION

Municipal Solid Waste (MSW) is a waste type which includes predominantly household waste with sometimes the addition of commercial wastes collected by a municipality within a given area. MSW is increasing in almost all part of the world. This is may be due to the population continues increasing in past ten decades. Bioreactor landfills are the most popular treatment system using for treating MSW at this time. Characteristics of MSW are different depending on its source and the people's lifestyles in that area. The Municipal Solid Waste management is shown in figure 1 and the characteristics of MSW are shown in figure 2.

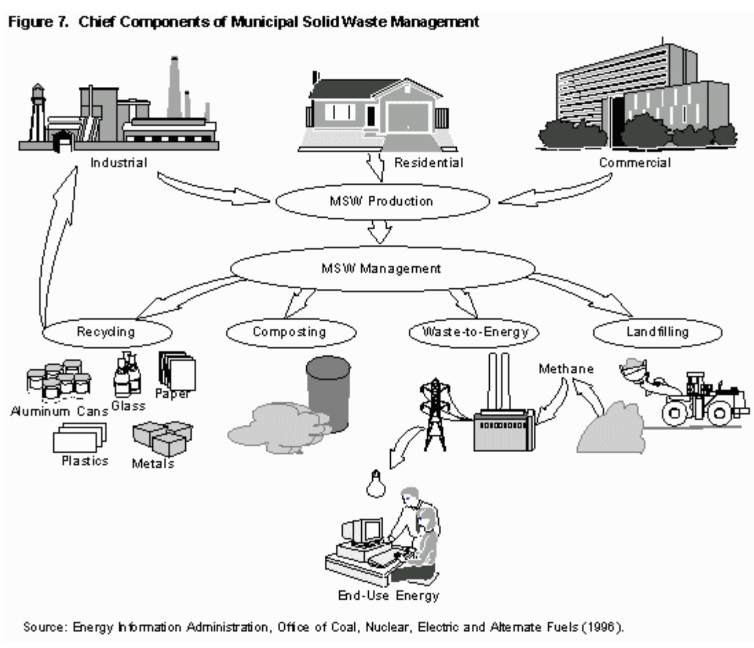


Figure 1 MSW management (http://www.eia.doe.gov)

PROJECTIONS OF MATERIALS GENERATED*
IN THE MUNICIPAL WASTE STREAM: 2000 AND 2005
(In thousands of tons and percent of total generation)

	Million tons		% of total	
Materials	**2000**	**2005**	**2000**	**2005**
Paper and Paperboard	87.7	94.8	39.3%	39.6%
Glass	11.9	11.2	5.3%	4.7%
Metals	17.6	18.7	7.9%	7.8%
Plastics	23.4	26.7	10.5%	11.2%
Wood	14.0	15.8	6.3%	6.6%
Others	19.7	22.2	8.8%	9.3%
Total Materials in Products	174.3	189.4	78.1%	79.1%
Other Wastes				
Food Wastes	22.5	23.5	10.1%	9.8%
Yard Trimmings	23.0	23.0	10.3%	9.6%
Miscellaneous Inorganic Wastes	3.4	3.6	1.5%	1.5%
Total Other Wastes	48.9	50.1	21.9%	20.9%
Total MSW Generated	223.2	239.5	100.0%	100.0%

* Generation before materials recovery or combustion.
Details may not add to totals due to rounding.
Source: Franklin Associates

Table 1 Characteristics of MSW in the United States, year 2000 and 2005. (www.epa.gov)

Leachate is the refuse from landfills and the compositions vary regarding the age of each landfills and the characteristics of MSW. Leachate is caused by the percolation of precipitation through MSW and react with the products of decomposition, chemical and other materials. Normally, leachate is anoxic, acidic, high COD, high sulfate and metal ion. The samples of leachate characteristics are shown below.

Table 6-2. Leachate Constituents of Conventional and Recirculating Landfil Summarizing all Phases

Parameter	Conventional*	Recirculating#
Iron, mg/l	20 - 2100	4 - 1095
BOD, mg/l	20 - 40,000	12 - 28,000
COD, mg/l	500 - 60,000	20 - 34,560
Ammonia, mg/l	30 - 3000	6 - 1850
Chloride, mg/l	100 - 5000	9 - 1884
Zinc, mg/l	6 - 370	0.1 - 66

*Pohland and Harper, 1986
#Natale and Anderson, 1985; Watson, 1993; Miller, et al, 1994; Lechner, et al, 1993

Table 2 Characteristics of leachate in conventional and recirculation landfills. (www.epa.gov)

ANAEROBIC DIGESTION

Anaerobic digestion is the reaction in which other compounds are used as the electron acceptor instead of oxygen. The reactions which could occur in anaerobic digestion of organic waste such as organic removal, denitrification, sulfide and sulfate removal are shown in figure 2.

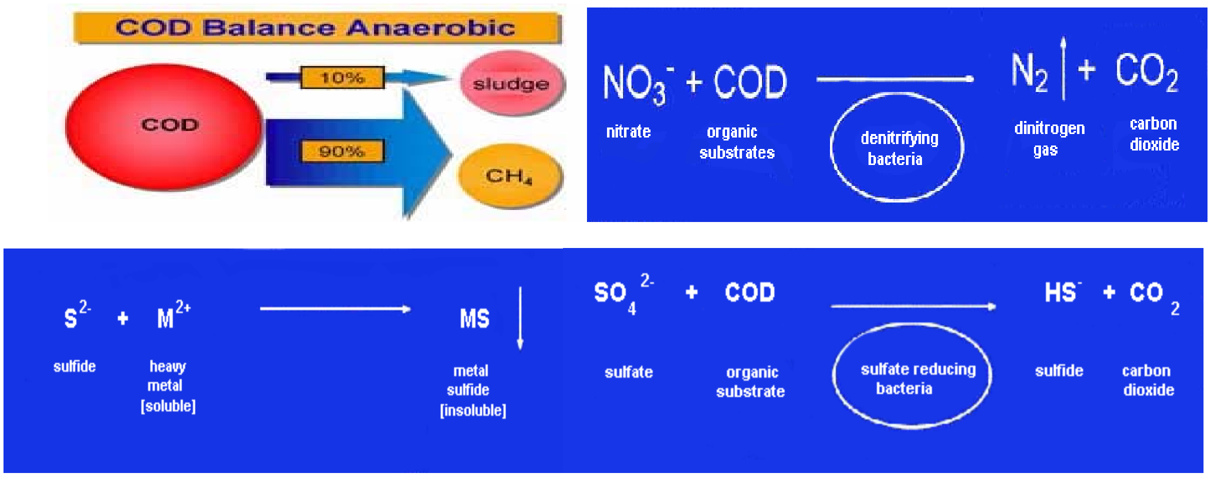


Figure 2 Reactions in anaerobic condition.

The transformation of complex compounds into biogas in anaerobic processes could be identified to four important stages as shown in figure 3.

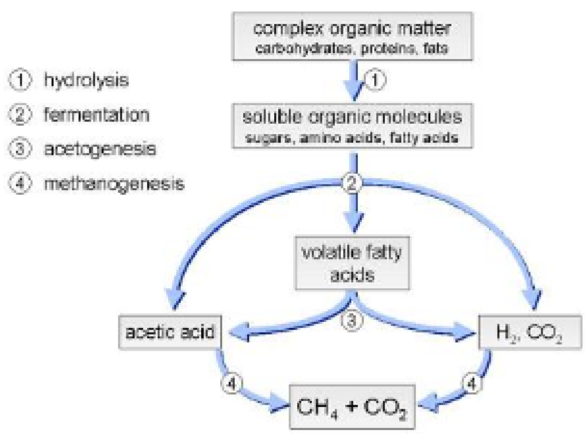


Figure 3 Four stages of organic transformation in anaerobic condition (www. water.me.vccs.edu).

1. Hydrolysis stage: Complex compounds are hydrolysed into dissolved compounds with a low molecular weight. Protein is degraded to aminoacid, carbohydrate is transformed to monosaccharides and disaccharides and lipid are converted to long-chain fatty acid.

2. Acidogenesis stage: The dissolved compounds are extracted to simple organic compound such as volatile fatty acid, alcohol, lactic acid and mineral compounds such as carbon hydroxide, hydrogen, ammonia and hydrogen sulfide.

3. Acetogenesis stage or fermentation: The products from acidogenesis stage are converted to suitable substrate of methanogen microorganism such as acetate, hydrogen and carbon dioxide.

4. Methanogenesis stage: This stage is often the limiting step of anaerobic digestion. In this step, carbon dioxide and hydrogen are converted to methane by hydrogenotrophic bacteria and acetate is converted to methane by methanogen bacteria (lettinga and Van Haandel,1990).

An anaerobic digestion technology is applied widely in landfills to treat MSW. The advantages of this technology are low consuming energy, producing landfill gas which could be utilized to renewable energy and high ability to treat high organic waste such as MSW. Leachate treatment is become the serious problem in almost all landfills all over the world. Because leachate with high toxic compound could contaminate ground water and cause the serious environmental problem. Recirculation is one of the most popular method to reduce leachate problem. Leachate recirculation could provide moisture to landfills and helps to enhance the efficiency of anaerobic digestion in term of reducing long-term environmental outcome, liability of waste storage and improving the economic of landfills (Reinhart and Townsend, 1997).

EFFECTS OF LEACHATE RECIRCULATION ON ORGANIC COMPOUNDS REMOVAL

Since, the boundary between each stage of an anaerobic digestion could not be determined exactly, COD, BOD and methane production are used to depict. The results from many studies in laboratory indicated that the lechate recirculation showed exact advantage on COD removal. The reactors with leachate recirculation showed significantly rapid degradation and also reached stabilization more quickly than those without recirculation. This might be because the lechate recirculation could decrease acidogenesis and methanogenesis period (Francois et al., 2007), (Chan et al., 2002). Moreover, the effects of leachate recirculation are clearly more effective in anaerobic reaction than those in aerobic reaction (Bilgili et al., 2007).

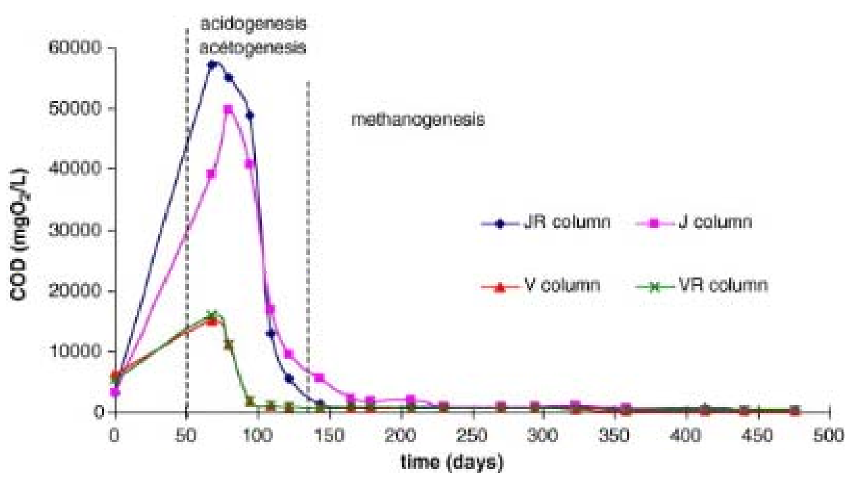


Figure 4 the effect of leachate recirculation on COD removal (Francois et al., 2007).

From figure 4, it was indicated that the leachate characteristics of bioreactor (JR,VR) and conventional landfills (J,V) is similar in term of COD. But in acidogenesis stage, COD concentration in leachate of bioreactor landfills was higher. It was possible that the dryness of conventional landfills and the poor contact between leachate and MSW make fewer leaching opportunity. The concentrations of volatile acid were different in leachate from landfills with and with out recirculation. In conventional landfills, the majority in volatile acid is butyrate but for bioreactor landfills, acetate has the highest concentration. pH value normally increase with decreasing of organic compounds in leachate. Since recirculated leachate had higher organic compounds when compared with single-pass leachate, it could be treated inside landfills and repeatedly recirculated back into the landfills until it reaches stabilization (Reinhart and Townsend, 1997).

The decreasing rate in COD is used for comparing between bioreactor and single-pass operation in order to consider the effect of leachate recirculation on leachate treatment. From decreasing rate, COD half-life of bioreactor landfills is shorter than those in conventional landfills. It means that bioreactor landfills reach stabilization stage more rapidly. Nevertheless, for full-scale bioreactor landfills, the COD half-life is about five times longer than those in laboratory because the overlapping phase occurs. Moreover, the recirculation devices in full-scale landfills are not as good as those being used in the laboratory. And also the compaction in full-scale landfills could effects leachate routing negatively. COD is removed from conventional landfills by washing out and biological conversion but in bioreactor landfills COD is mainly removed by accelerated biological conversion (Reinhart and Townsend, 1997).

Another conclusion is the increasing of the moisture content of waste by recirculation of leachate has improved the leaching process and biochemical reactions within MSW in landfills. However, the waste in conventional landfills receives moisture only from rainfall. The higher pH, which might reflect a lost of volatile acid, would also aid development of methanogen bacteria. The rapid development of methanogen bacteria in bioreactor landfill is the result from high moisture content (Christensen et al., 1992).

However, in some cases, the reactors with leachate recirculation reached stabilization after those without recirculation. This may be due to the leakage of methanogen bacteria causing by leachate washing out. The over dose of leachate recirculation could effect in this situation (Sanphoti et al., 2006), (Sponza et al., 2004).

EFFECT OF LEACHATE RECIRCULATION ON LANDFILL GAS PRODUCTION AND METHANE CONTENT

The cumulative volume of methane and carbon dioxide production increased with leachate recirculation for about 1.7 to 2 times when compared with the reactors without leachate recirculation. However, the higher recirculation rate may cause the decreasing in cumulative methane production. It was possible that acidic conditions could inhibit the methanogenesis bacteria activity (Šan et al., 2001).

Table 6

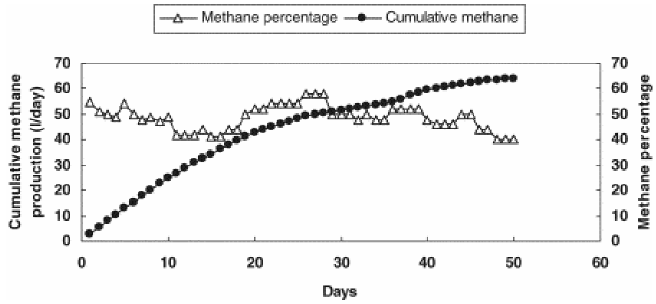
CO_2 and CH_4 of biogas percentages released by J, JR, V and VR columns over time

	At the end of 50 days of recirculation		At the end of 125 days of recirculation		>200 days of recirculation		Vtotal of CO_2 (L/kg of waste)	Vtotal of CH_4 (L/kg of waste)
	% CO_2	% CH_4	% CO_2	% CH_4	% CO_2	% CH_4		
JR column	80	20	53	47	54	46	37.7	32.3
J column	85	15	57	43	54	46	16.5	14.5
V column	46	54	45	55	47	53	12.7	14.3
VR column	45	55	46	54	46	54	21.1	24.9

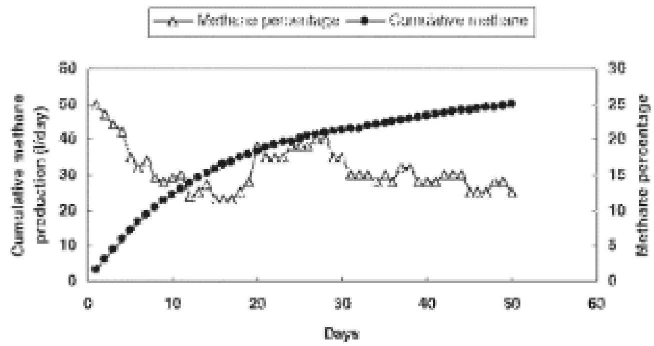
Table 3 CO_2 and CH_4 of biogas percentages released (Francois et al., 2007).

The proportion of methane in landfill gas is distinctly higher in recirculated reactors than those without recirculation (Sponza et al., 2004), (Šan et al., 2001). It is possible that the recirculation could increase methanogenesis bacteria activity. The leachate recirculation also could shorten the methanogenesis stage when MSW is co-digested with sewage sludge and marine dreading (9 weeks and 11 weeks respectively). Nevertheless, the gas production depends on some factors such as pH, temperature, alkalinity and absence of toxic compound (Chan et al., 2002). Moreover, the increasing in gas generation has a relationship with the higher degree of stabilization and adding buffer solution before recirculation might accelerate methanogenic bacteria in waste degradation and could enhance gas production rate (Šan et al., 2001).

The gas production is more difficult to measure in full-scale landfills when compared with laboratory scale. The landfill gas production is enhanced by both accelerated COD removal rate, the return of organic compounds and methanogen bacteria. In conventional landfills, the organic matter is washed out by single-pass leachate. For example, the data from parallel 2.5 ha cells operated by the Delaware Solid Waste Authority showed that the gas production increase ten times when compared with conventional cell. Moreover, the researchers from the University of Central Florida in Orlando indicated that the gas production rate was 59% higher in bioreactor landfills when compared with conventional landfills (Reinhart and Townsend, 1997).



(a)



(b)

Figure 5 Quantity of methane and methane percentage of reactor with (a) and without leachate recirculation (b) (Sponza et al., 2004).

EFFECT OF LEACHATE RECIRCULATION ON METAL CONCENTRATIONS

An effect of leachate recirculation on metal concentrations of a simulated landfill was studied by Bilgili et al. (2007). The results show that the concentrations of metal decrease about 50% in 250 days after increasing at the beginning of the experiment. This was because at that period pH of leachate is low (4.0–6.0) in the acidogenensis stage, so the metals had high solubility and dissolve into leachate. After reaching methanogenesis stage, pH was increased to neutral and the metal could precipitate and were trapped in MSW as shown in figure 6 and 7.

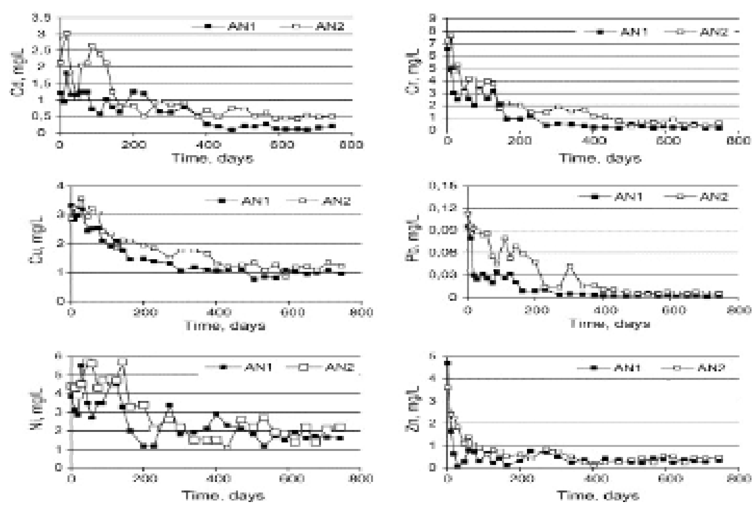


Figure 6 The changes in Cd, Cr, Cu, Pb, Ni, and Zn concentrations in anaerobic landfill leachate (Bilgili et al., 2007).

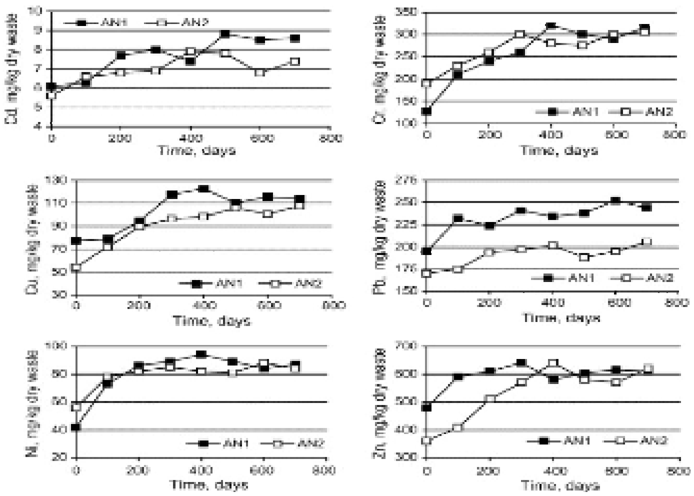


Figure 7 The changes in Cd, Cr, Cu, Pb, Ni, and Zn concentrations in waste samples (Bilgili et al., 2007).

pH is the important parameter of the solubility ability of metal. The pH that make the metals have minimum solubility is about 7.0 – 10.0. The precipitation might be the precipitation of metal with sulfides, carbonate and hydroxide (Bilgili et al., 2007).

Another example is at closed bioreactor sites, iron concentration tend to decrease with time but still constant in conventional landfills. The primary mechanisms of metal removal of bioreactor landfills appear to be metal sulfide and metal hydroxide precipitation and subsequent capture with in the waste metrix by encapsulation, sorption, ion-exchange and filtration. Gould et al. (1989) found that leachate recirculation simulated reducing condition in lysimeters, providing for reduction of sulfate to sulfide which causes low concentration of leachate. Chain and Dewalle (1977) reported that the formation of metal sulfide under anaerobic condition effectively eliminated the majority of heavy metal in leachate. Moreover, metal hydroxide precipitation is enhanced under neutral or above neutral leachate condition which are stimulated by leachate recirculation. In addition, high molecular weight humic-like substances are formed and tend to form complex with heavy metal (Reinhart and Townsend, 1997).

THE STUDIES OF LEACHATE RECIRCULATION IN FULL-SCALE LANDFILLS

The long term monitoring studies at MSW landfill facility with leachate recirculation was studied by Morris et al. (2003). These landfills are located in the Central Solid waste Management Center, Delaware, USA. These sites have been operated since 1980 with the opening of 3.6 ha. (area A) after that, the second landfill was installed with area about 7.3 ha next to area A. Both of them are operated with leachate recirculation with (1) leachate injection well, (2) surface spray and (3) top surface application via leachate recirculation fields. Moreover, two test cells were installed and operated with and without leachate recirculation to observe the difference between them.

Properties of landfill facilities

	Area A/B	Cell 1	Cell 2
Facility opened	October 1980	August 1989	August 1989
Facility closed	October 1988	October 1996	October 1996
Area (ha)	10.9	0.4	0.4
Waste mass (tonnes)	642,290	7900	8300
Total volume (m^3)	1,287,900	16,000	16,000
Cover soil (% by volume)	23	29	27
Period of leachate recirculation	1985–1995 (years 7–17)	1990–1996 (years 2–8)	NA
Total volume of leachate recirculated	72,000 m^3	1920 m^3	NA

Table 4 properties of landfill facilities (Morris et al., 2003).

The findings of this research in A/B full scale landfills show that the volume of leachate generated did not increase significantly during recirculated period. The large volume of rainfall tends to had higher influence.

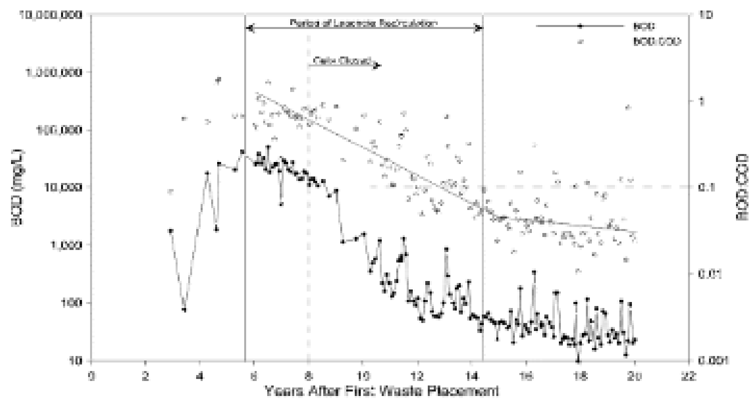


Figure 8 BOD concentration and BOD:COD ratio in Area A/B leachate (Morris et al., 2003).

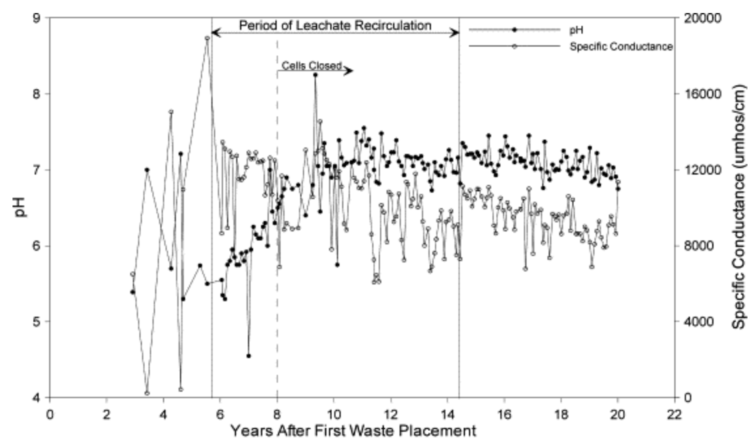


Figure 9 pH and conductivity of lechate from Area A/B (Morris et al., 2003).

And both rain water infiltration and lechate recirculation provide the positive effects to increase moisture content. The concentrations of pH, BOD and BOD:COD showed that this landfill reach methanogenesis stage in around year 6. However, figure 10 showed that ammonia nitrogen concentration in leachate maintained about 300 – 400 mg/L and tended to decrease in year 15. This may be because of the rate of flushing experienced by the landfill (Morris et al., 2003).

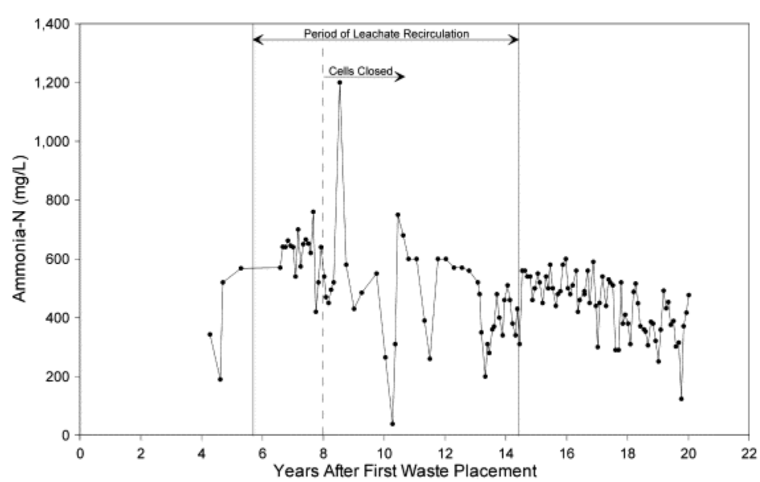


Figure 10 Ammonia concentration in Area A/B leachate (Morris et al., 2003).

Information from test cell:
The pH data from cell 1 with leachate recirculation showed that this cell reached neutrality about year 3 and remained stable in the rest of observed period. On the other hand, the pH data of cell 2 without leachate recirculation showed inconsistent data and remained under neutrality along observed period and moreover, it tended to decline after year 6. It means that the cell 1 reached methanogennesis stage very soon after the first recirculation of leachate and cell 2 has never reached this stage.
Nevertheless, it might be described that the rapid increasing in pH after leachate recirculation in cell 1 might due to the small contact proportion between leachate and MSW. BOD concentration and BOD/COD ratio did not have significantly difference during experimental period. Ammonia concentration in cell 1 is higher than cell 2 and tends to increase after 3.5 years because of the cumulative of ammonia from recirculated leachate (Morris et al., 2003).

The data clearly indicated that the leachate recirculation could enhance the landfill gas production. The quantity of landfill gas production of cell 1 is about 10 times higher than cell 2 during experimental period. The evidence that support the more rapid degradation of the landfill with leachate recirculation compared with those without recirculation is shown in this experiment. The researchers of this study buried time capsules which contained chicken legs in each cell and observed them more than 8 years later. Figure 11 shows that the chicken leg from cell 1 has been degraded to a fragile bone but the chicken leg in cell 2 is still largely intact (Morris et al., 2003).

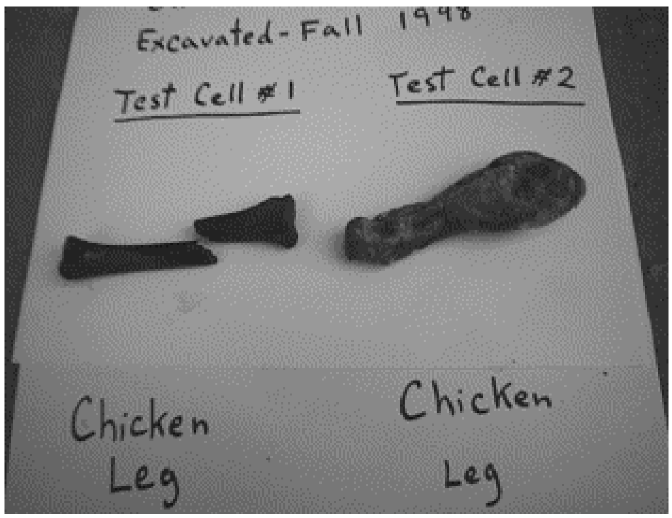


Figure 11 Selected results from the time capsule study performed at the test cells (Morris et al., 2003).

Benson et al. (2007) studied about the effect of leachate recirculation in five landfills in North America. The data show that leachate generation rate from both with and without landfills are not significantly different in the experimental period (May 02 to Oct. 02). It is possible that the waste is below absorption capacity and can absorb recirculated leachate. However, the effect of rainfall precipitation could affect the quantity of leachate as well.

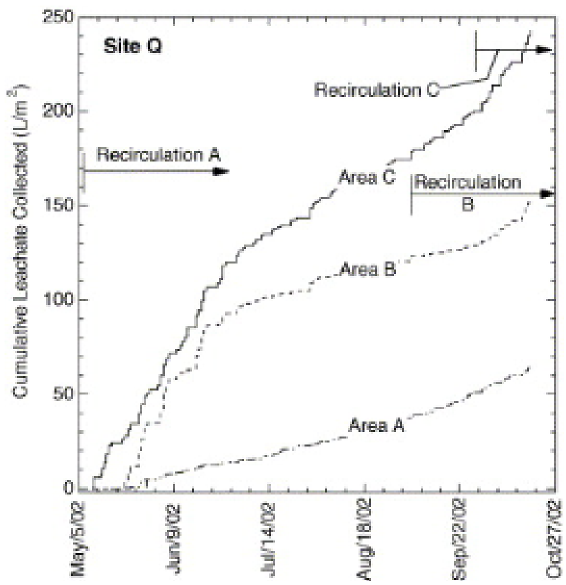


Figure 12 Cumulative leachate collected per unit area at Landfill Q before and after recirculation was initiated (Benson et al., 2007).

The landfill gas production rate measuring in term of gas flow per unit length of well screen showed that the gas flow rate from bioreactor landfill was 69% higher than landfill without leachate recirculation as shown in figure 13.

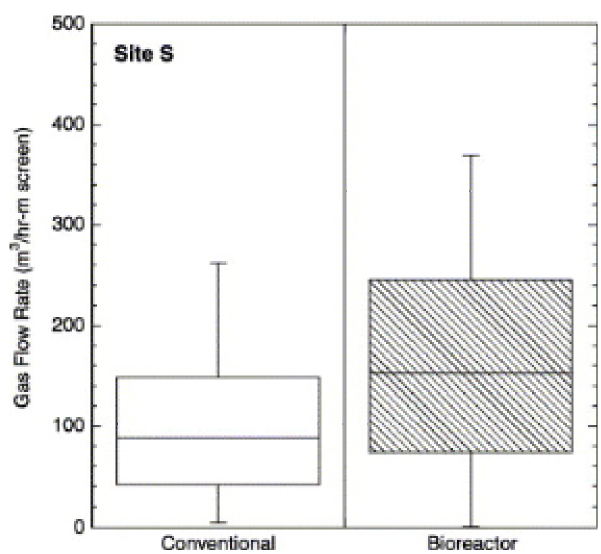


Figure 13 Box plots showing gas flow rates per unit length of well screen from conventional and bioreactor landfills at Landfill S (Benson et al., 2007).

The characteristics of leachate were studied from year 1992 to 2002 in term of pH, BOD, BOD:COD and ammonia concentration. The results indicated that pH tended to increase for about one year after leachate was recirculated because of the stimulation of hydrolytic and fermentative bacteria. After that, pH increased and become stable between 7.0 and 8.0 because this landfill reached methanogenesis stage. The COD and BOD increased in the first 2 years after recirculation and became decreasing in year 3. This is due to the accumulation of carboxylic acid. The BOD:COD ratio which show the degradable organic compounds varied from 0.5 – 0.7 before recirculation. After that, this ratio decreased slightly in the first 3 years of recirculation and reached about 0.1 which showed the well decomposed MSW in year 4 (Benson et al., 2007).

Ammonia-nitrogen concentration increased with the increasing of leachate recirculation rate because of the absence of nitrification microorganism which works effectively in aerobic condition. From this reason, ammonia-nitrogen tends to accumulate in these landfills (Benson et al., 2007).

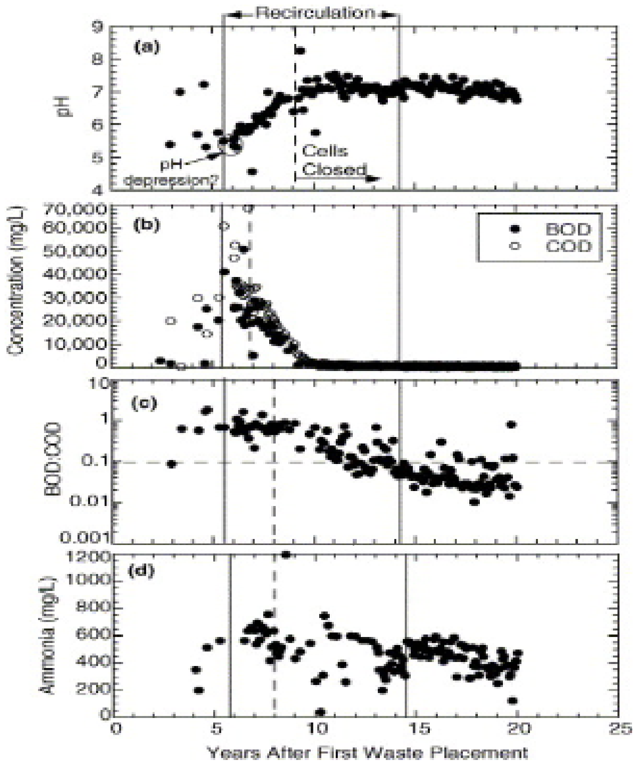


Figure 14 Leachate quality variables for Landfill C as a function of time: (a) pH, (b) BOD and COD, (c) BOD:COD ratio, and (d) ammonia concentration (Benson et al., 2007).

Leachate volume of full-scale landfills increases with the increasing leachate recirculation rate. Because of the current good capping technique, recirculation leachate volume tends to be dominant after landfill closure. The data from six full-scale landfills show that leachate generation vary from 1.1 to 13.5 m^3/ha/d with recirculation rate about 40 to 70% of leachate generated. In the other hand, for conventional landfills, leachate generation is depending on climate and site characteristics and off-site disposal leachate vary from 0 to 59% of leachate generated. For bioreactor landfills, the volume of leachate treatment depends on both volume of leachate generated and available storage volume. Unlike conventional landfill, if storage volume is small, the recirculating rate tends to be higher than those with large storage volume (Reinhart and Townsend, 1997).

DISADVENTAGES OF LECHATE RECIRCULATION

Although, leachate recirculation tended to make positive effects to engineering landfills, Disadvantages are reported by some researchers. The accumulation of chloride ion occurs when leachate was circulated so leachate recirculation seems to concentrate this pollutant rather than remove it. To prevent this problem, leaching out or the flushing reactor might be the most effective process (Francois et al., 2007).

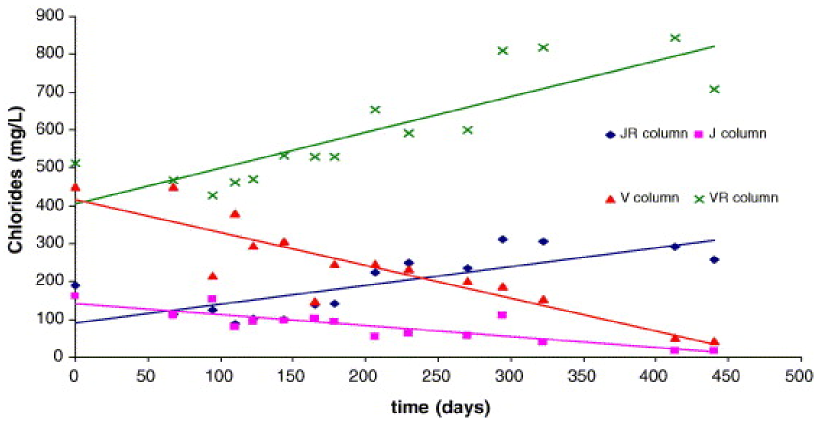


Figure 15 Development of chloride content in leachates resulting from the columns with (JR, VR) and without leachate recirculation (J, V) (Francois et al., 2007).

And also, as shown in figure 10 and 14, ammonia will be accumulated in the landfills. Ammonia concentration about 1,500 to 3,000 mg/L will damage methanogen bacteria. To eliminate this problem, leachate should be pre-aerated before recirculation (Francois et al., 2007).

Moreover, leachate recirculation might not be appropriate in some areas such as UK because of this following reason. The rainfall exceeds potential evaporation and evatranspiration so the volume of leachate available for recirculation is greater than requirement and need to be discharged and treat off-site (Christensen et al., 1992).

RECIRCULATION STRATEGIES

The recirculation strategies are shown by Reinhart and Townsend, (1997). The most important key point of leachate recirculation is "the balance of biological process of waste degradation and the hydrologic capacity of the waste which controlled by the waste permeability". Landfill process control could be improved by controlling the recirculation rate. The strategy of leachate recirculation is to recycle leachate slowly before the landfills reach methanogenesis stage. High flow rate could remove microorganism and decrease buffering capacity. The methane production shows that methanogenesis stage occurring. Acid stuck conditions are indicated by a high volatile acid to alkalinity ratio (greater than 0.25) since pH might still in acceptable range, the process may be inhibited.
Leachate recirculation should be initiated as soon as possible following waste placement to ensure the appropriate moisture content of biodegradation. When landfills reach stabilization, leachate could be recirculated more frequency and higher flow rate. At this point, the rate of inauguration of leachate is controlled by the moisture capacity of the waste.
Another important strategy is the leachate should be recirculated to all part of landfills. Non-uniform distribution could lead to short circuiting which could reduce gas production rate. With time, the density of the waste in landfill will increase so the acceptable of leachate recirculation rate decrease as well.

CONCLUSION

Since leachate is one of the important problems in engineering landfills process because it has high concentration of toxic compound and could contaminate ground water. The leachate recirculation is one of the suitable methods to solve these problems. The recirculation shows a lot of benefits on anaerobic landfills as shown below.

- Accelerating the degradation
- Increase in Landfill gas production
- Reduce metal concentration in leachate

However, there are several disadvantages of the leachate recirculation such as the accumulation of ammonia and chloride and the climate of each area. From those pros and cons, the leachate recirculation tends to be one of the most appropriate methods to apply in anaerobic landfills. To reach the optimum benefit of leachate recirculation, the designers and operators of bioreactor landfills should balance hydraulic and biological pattern properly.

REFERENCES

Benson, C. H.; Barlaz, M. A.; Lane, D. T.; and Rawe, J. M., B.E. (2007) Practice review of five bioreactor/recirculation landfills. *Waste Management*, **27**,13-29

Bilgili, M. S.; Demir, A.; and Özkaya, B., B.E. (2007) Influence of leachate recirculation on aerobic and anaerobic decomposition of solid wastes. *Journal of Hazardous Materials*, **143**, 177-183

Bilgili, M. S.; Demir, A.; İnce, A.; and Özkaya, B., B.E. (2007) Metal concentrations of simulated aerobic and anaerobic pilot scale landfill reactors. *Journal of Hazardous Materials*, **145**, 186-194

Chan, G. Y. S.; Chu, L. M.; and Wong, M. H., B.E. (2002) Effects of leachate recirculation on biogas production from landfill co-disposal of municipal solid waste, sewage sludge and marine sediment. *Environmental Pollution*, **118**, 393-399

Chian, E. S. K., B.E. (1977) Stability of organic matter in landfill leachates. *Water Resource*, **11(2)**, 159

Christensen, T. H.; Cossu, R.; and Stegmann, B.E. (1992) Landfilling of waste: leachate; Elsevier Applied Science: London and New York.

Francois, V.; Feuillade, G.; Matejka, G.; Lagier, T.; and Skhiri, N., B.E. (2007) Leachate recirculation effects on waste degradation: Study on columns. *Waste management*, **27**, 1259-1272

Gould, J. P.; Cross, W. H.; and Pohland, F. G., B.E. (1989) Factors influencing mobility of toxic metals in landfills operated with leachate recycle. In *Emerging Technologies in Hazardous Waste Management*, *D. W. Tedder and F. G. Pohland, Ed., ACS Symposium Series 422*(1989).

Lettinga, G.; and Van Haandel, A. C., B.E. (1994) Anaerobic Sewage Treatment; John Wiley & Sons: New York.

Morris, J. W. F.; Vasuki, N. C.; Baker, J. A.; and Pendleton, C.H., B.E. (2003) Findings from long-term monitoring studies at MSW landfill facilities with leachate recirculation. *Waste Management*, **23**, 653-666

Reinhart, D. R.; and Townsend, T. G., B.E. (1997) Landfill Bioreactor Design & Operation; Lewis Publishers: New York.

Šan, I.; and Onay, T. T., B.E. (2001) Impact of various leachate recirculation regimes on municipal solid waste degradation. *Journal of Hazardous Materials*, **87**, 259-271

Sanphoti, N.; Towprayoon, S.; Chaiprasert, P.; and Nopharatana, A., B.E. (2006) The effects of leachate recirculation with supplemental water addition on methane production and waste decomposition in a simulated tropical landfill. *Journal of Environmental Management*, **81**, 27-35

Sponza, D. T.; and Ağdağ O. N., B.E. (2004) Impact of leachate recirculation and recirculation volume on stabilization of municipal solid wastes in simulated anaerobic bioreactors. *Process Biochemistry*, **39**, 2157-2165