

## Nitrogen removal during leachate treatment: comparison of simple and sophisticated systems

J.-L. Vassel\* and H. Jupsin\* and A.P. Annachatre\*\*

\* Fondation Universitaire Luxembourgeoise, Arlon, Belgium (E-mail: [vassel@ful.ac.be](mailto:vassel@ful.ac.be); [jupsin@ful.ac.be](mailto:jupsin@ful.ac.be))

\*\* Environmental Engineering and Management, Asian Institute of Technology, PO Box 4, Klongluang, Pathumthani, 12120 Thailand (E-mail: [ajit@ait.ac.th](mailto:ajit@ait.ac.th))

**Abstract** Membrane bioreactors (MBR) have become common in treating municipal wastewaters. Applied to leachates treatment MBR were also successful with pilot scale experiments and full-scale facilities as well. We succeeded previously in designing an efficient nitrification–denitrification process with an ethylene glycol byproduct as carbon source for denitrification. Moreover, an unexpectedly high inert COD removal efficiency was also observed in the full-scale MBR facility thereby making it possible to increase the operating time of the final GAC (Granulated Activated Carbon) adsorber. Since MBR are very sophisticated systems. Simpler and “lower” cost systems can also be considered. For example it is possible to nitrify leachates from sanitary landfill using a simple infiltration–percolation technique with a low energy cost. To validate previously published laboratory experiments, a semi industrial-scale pilot installation was installed at the Montzen landfill site (Belgium). The process is based on infiltration–percolation through a granular bed. This well known process was modified to increase the load, notably by changing the support medium, adding an electric fan that is run intermittently and maintaining temperatures greater than 15°C. The new material is a type of granular calcium carbonate with a large specific surface area. These technical improvements enabled the system to nitrify up to 0.4 kg NH<sub>4</sub><sup>+</sup>-N/m<sup>3</sup> of reactor bed per day at a hydraulic load of 0.35 m.d<sup>-1</sup>, with an ammonia removal rate in the range of 80 to 95%. Despite the high ammonia nitrogen inlet concentrations, this system exhibits remarkable nitrification efficiency. Moreover, these performances are achieved in a batch mode system without recirculation or dilution processes. If complete nitrification is needed, it can be obtained in a second in series of bioreactors. The system can be classified as a low cost process. An international patent is pending. Possible performances of those systems were compared with the usual methods for leachates treatment.

**Keywords** Denitrification; leachate; MBR; nitrification

### Introduction

Biological treatment of old leachates is a difficult problem as the largest part of COD is non-biodegradable and a huge concentration of N-NH<sub>4</sub><sup>+</sup> is still present. This means that conditions for nitrification have to be set up but depending also on the national regulations for denitrification. In the latter case, if the leachates are “old” which means that biodegradable COD is not available, an external carbon source is needed. Many different types of system have been used for leachates treatment (Rees, 1982) such as activated sludge, trickling filters and even more extensive systems. Among them, membrane bioreactors (MBRs) have become a common practice to treat municipal sanitary landfill (MSL) leachates (Bressi and Favali, 1997; Luning and Notenboom, 1997; Praet *et al.*, 2000, 2002; Wens *et al.*, 2001). The performance of a MBR treating leachates from an old MSL that has now entered the aftercare period was monitored for three years. The typical characteristics of such leachates include rather high C and N concentrations (COD = 3,000 mgO<sub>2</sub>/l; [NH<sub>4</sub><sup>+</sup>] = 1,200 mg N/l) combined with poor biodegradability (BOD<sub>5</sub>/COD < 10%). In this study, the treatment has been tested first by means of pilot scale experiments then a full-scale facility has been designed, started up and operated (Figure 1). Results confirm high nitrogen removal efficiency in MBR. Additionally the MBR removed a significant proportion of the

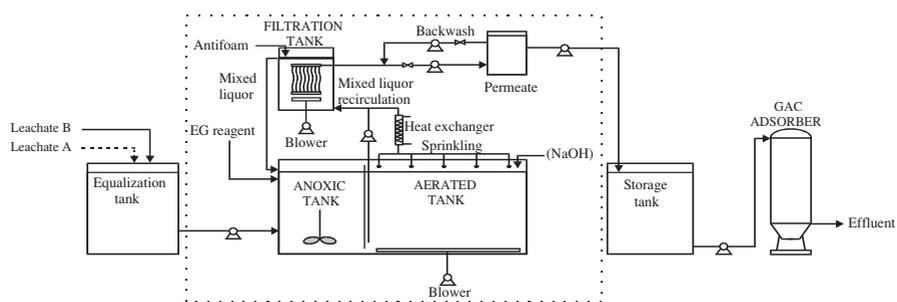
soluble refractory COD, thereby making it possible to increase the operating time of the final GAC adsorber. The bioreactor has been described elsewhere (Praet *et al.*, 2001). In a nutshell, it consists of three main compartments, namely,

1. a 70 m<sup>3</sup> aerated compartment to nitrify N-NH<sub>4</sub><sup>+</sup> nitrogen;
2. a 30 m<sup>3</sup> anoxic stirred-tank compartment in which N-NO<sub>3</sub> is denitrified; and
3. a 10 m<sup>3</sup> aerated compartment in which the filtration *per se* takes place.

Given the low BOD/N ratio, an efficient biodegradable exogenous carbon source (monoethylene glycol) is added as a carbon source for denitrification (Praet *et al.*, 2000). Finally, an activated carbon adsorber is used to remove the remaining COD. As indicated in Table 1, there are some differences between the full- and pilot-scale facilities, but most of the results obtained in the preliminary study were confirmed (Praet *et al.*, 2002).

### Main results on MBR

Very efficient nitrogen removal was obtained for leachate treatment in a MBR. Old leachates require an exogenous carbon source if denitrification is desired. In this study a very efficient and inexpensive carbon source composed mainly of ethylene glycol was utilised. Results showed greater than 96% nitrogen removal efficiencies during the last two



**Figure 1** Schematic diagram of the treatment plant

**Table 1** Main characteristics of the pilot MBR and of the full scale facility

	Pilot	Full scale facility
Total working volume (m <sup>3</sup> )	5	100
Anoxic tank volume (m <sup>3</sup> )	2.5	27
Aerated tank(s) volume (m <sup>3</sup> )	2.5	73
Aerated volume (%)	50	73
Membrane area (m <sup>2</sup> )	17	Initially: 92 Now: 138
DO concentration (mgO <sub>2</sub> /l)	≅ 1.5	≅ 1.5
Work cycle	200 sec. suction 50 sec. backwash 200 sec. rest	360 sec. suction 60 sec. backwash
Air flow rate (m <sup>3</sup> /h), fine bubbles	18–21	600
Membrane aeration (m <sup>3</sup> /h), coarse bubbles	18–24	200
Recycle flow rate (m <sup>3</sup> /h)	2.5	25
Inlet flow rate (m <sup>3</sup> /d)	1.25	25
Nominal permeate flux rate (l.m <sup>-2</sup> .h <sup>-1</sup> )	3.1	7.5
Average hydraulic residence time (d)	4	4
[NH <sub>4</sub> -N] (mg/l)	1,200	650–750
[MLVSS] (g/l)	15	15
NH <sub>4</sub> -N loading rate (kgN/d)	1.5	16.25 to 18.75
Exogenous carbon for denitrification	Methanol	Ethylene glycol
(COD/N ratio used)	(>5)	(≅ 4–5)

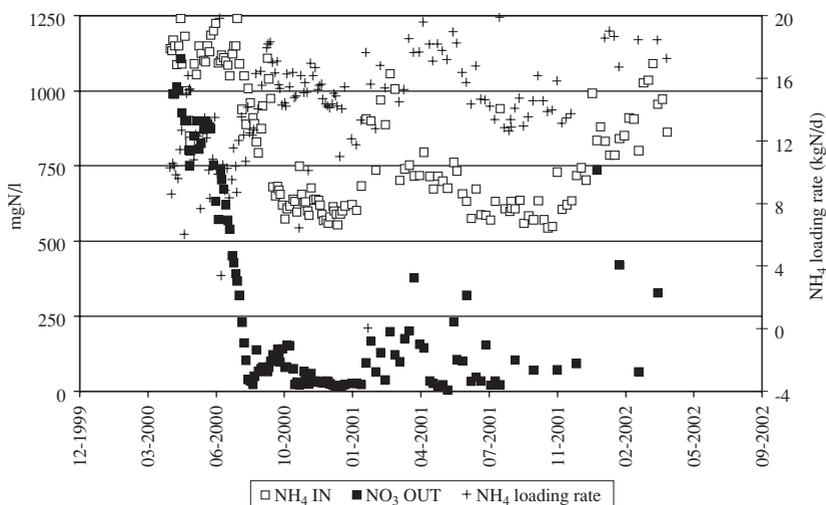
months of the follow-up (mean total inorganic nitrogen concentration: 29 mgN/l). Moreover no pH control (alkalinity addition) is needed in the nitrification denitrification process. In this study, the short membrane lifetime of the MBR (< 1 year) was found to be the main disadvantage of the system but no problem occurred since the total membrane area has been increased. Most of the particular behaviours revealed during this study for MBR processes (suction pressure, membrane lifetime, specific activities, oxygen transfer, etc.) demonstrate the need for intensive and long-lived preliminary pilot studies with the leachates to be treated. The COD/N and  $V_{\text{anoxic}}/V_{\text{aerated}}$  ratios have still to be optimised. Surprisingly, a large removal of soluble refractory COD was observed in the MBR, leading to a large saving in GAC consumption. The precise mechanisms of this phenomenon are still under study: due to the very high sludge age obtained in MBRs, microorganisms able to degrade refractory COD compounds such as humic acids are able to grow in the reactor. Such microorganisms have been isolated and are now being identified. They will ultimately be patented. Their specific activities are rather high, which may open possibilities for much higher refractory COD removal efficiency in leachates treatment. COD removal is a major economic advantage of this process because it can drastically reduce the cost of the activated carbon used to adsorb the inert COD (Jupsin *et al.*, 2003).

### Infiltration-percolation system

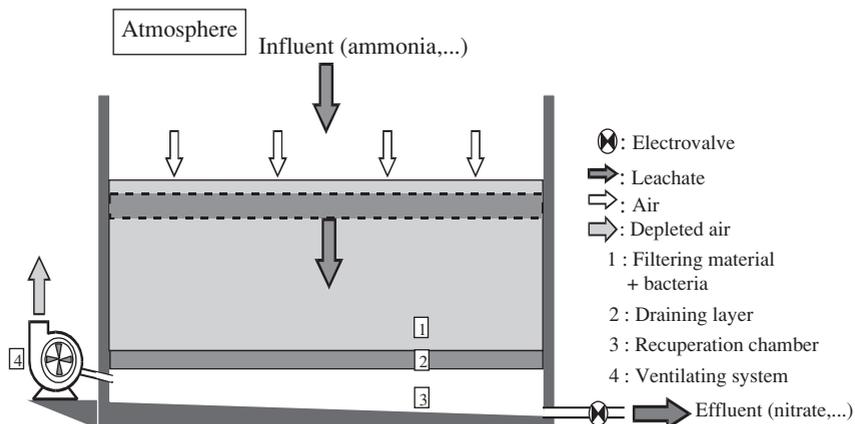
In another study a low cost system that may reduce ammonia nitrogen pollution was validated. This system was first developed at the laboratory scale on small columns. Effectively, the lab scale study focused on the choice of an appropriate media, hydraulic loading, management and the air renewal and ventilation management.

### Description of the experimental device and operating conditions

The semi-industrial-scale pilot experimental setup is a 2 m HDPE height cylindrical reactor with a 4.3 m<sup>2</sup> horizontal section. One metre of garnishing material (lithothamnium) lies on a 15 cm gravel-draining layer (Figure 3). A fan is connected to a water collection chamber where it draws fresh air through the top of the infiltration surface. The feed tank is equipped with an overflow pipe that permits control of influent volume at each feeding period, thus also controlling hydraulic loading. Once in the collection chamber, the treated leachate is evacuated by a centrifugal pump connected to an electrovalve (Figure 3).



**Figure 2** Nitrogen removal efficiencies

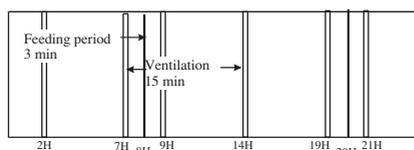


**Figure 3** Principle of the infiltration–percolation process for leachates treatment (Nitrification)

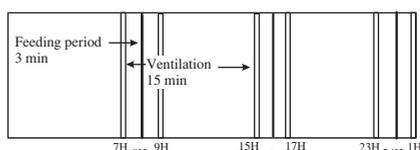
The operating conditions (Figures 4a and 4b) selected during the study are summarized in Table 2. The remainder of this paper will be focused on leachates described in Table 3. To prevent damage from freezing, the installation was built in a greenhouse and kept at a temperature of 20°C. Given the thermal inertia of the filtration bed, the low temperature of the influent (at the beginning of the study) and the sensitivity of nitrification processes to temperature, the variations of the temperature during the first part of this study using a temperature probe located in the reactor at 30 cm depth were recorded continuously.

### Results and comments on the infiltration–percolation process

The nitrification results are presented in Table 4. Ammonia concentrations at the outlet of the prototype fluctuated between a minimum value of 7 mg NH<sub>4</sub><sup>+</sup>-N/l and a maximum of 900 mg NH<sub>4</sub><sup>+</sup>-N/l (Figure 5). At the rate of 0.35 m.d<sup>-1</sup> the system oxidized an average of 325 g N.m<sup>-3</sup>.d<sup>-1</sup>, and 410 g N.m<sup>-3</sup>.d<sup>-1</sup> at 0.52 m.d<sup>-1</sup> (corresponding to a removal rate of 79.8% and 72.5% respectively). However, the system’s performance degraded at higher hydraulic loading and decreased to 40% by the end of the study. As nitrification processes are highly influenced by temperature, a heating system was provided in the feeding tank. Raising the temperature to 20–25°C, led to a marked improvement in nitrification. In fact, at 0.35 m.d<sup>-1</sup>, removal varied from 80 to about 100% when the temperature was above 20–25°C but dropped to around 50% at 10°C. However, a hysteresis effect was noticed on the growth of nitrifiers when the temperature was changing. At the end of the winter season,



**Figure 4a** Management of feeding and ventilating periods at the rate of 0.35 m.d<sup>-1</sup>



**Figure 4b** Management of feeding and ventilating periods at the rate of 0.52 m.d<sup>-1</sup>

**Table 2** Operating conditions tested on the pilot

Hydraulic rate m.d <sup>-1</sup>	Feed cycles per day	Nitrogen loading g NH <sub>4</sub> <sup>+</sup> -N.m <sup>-3</sup> .d <sup>-1</sup>	Ventilation frequency	Duration days
0.35	2	408	6 × 15 min per day	143
0.52	3	565	6 × 15 min per day	62

heating was stopped in the feed tank, nitrification decreased slightly and progressively to reach a minimum of 75%; at that time the temperature was around 15–17°C. After this period the air temperature increased and the influent temperature increased as well to around 20–25°C, after which some removal efficiencies were recorded in the range of 92% to 97% (yielding 0.4 kg.N-NH<sub>4</sub><sup>+</sup> m<sup>-3</sup> of reactor. d<sup>-1</sup>).

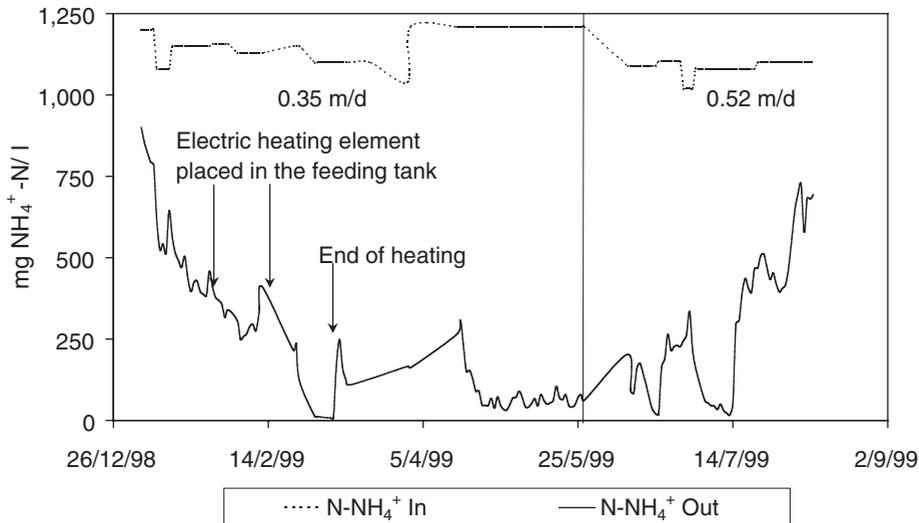
At steady state, nitrification performances at 0.35 m.d<sup>-1</sup> were obtained and the hydraulic load was increased to 0.52 m.d<sup>-1</sup> (divided into three applications per day). At the higher loading, NH<sub>4</sub><sup>+</sup>-N at the outlet of the system increased progressively until the removal at the end of the study was only 40% (Figure 5).

**Table 3** Characteristics of the leachates emitted by the Eastern area of Montzen landfill (1998)

Parameters	NH <sub>4</sub> <sup>+</sup> mg N/l	NO <sub>2</sub> <sup>-</sup> +NO <sub>3</sub> <sup>-</sup> mg N/l	Alkalinity meq/l	Unfiltered BOD <sub>5</sub> mgO <sub>2</sub> /l	Unfiltered COD mgO <sub>2</sub> /l	pH	Conductivity µS/cm
Mean	1,274	0.05	120	223	3,040	7.56	14,960
Std deviation	60	0.04	9	21	243	0.16	1,089
Maximum	1,365	0.06	128	240	3,880	8.1	17,540
Minimum	967	0.01	115	200	2,350	7.3	12,490
Number of observations	52	6	6	3	51	52	52

**Table 4** Nitrification efficiencies obtained on the pilot device set up at Montzen landfill

	Influent	Effluent	
		Q = 0.35 m.d <sup>-1</sup>	Q = 0.52 m.d <sup>-1</sup>
pH	7.6	8	8.1
Conductivity (µS.cm <sup>-1</sup> )	15,160	13,306	12,690
Ammonia concentration (mg NH <sub>4</sub> <sup>+</sup> -N/l)	1,126	235	298
Standard deviation	54	220	222
Efficiency (gN.m <sup>-3</sup> .d <sup>-1</sup> )	–	325.8	410
Removal rate (%)	–	79.8	72.5
Number of observations	161	98	63



**Figure 5** NH<sub>4</sub><sup>+</sup>-N concentration at inlet and outlet of the reactor under at two hydraulic rates

### Nitrogen balance

Various authors have observed that more than 85% of the total nitrogen in sanitary landfill leachates is ammonia (Robinson *et al.*, 1982; Vassel *et al.*, 1991; Imai *et al.*, 1993). As  $\text{NH}_4^+\text{-N}$  represents about 95% of Kjeldahl nitrogen in the present study, the nitrogen balance on the basis of ammonia nitrogen removed compared to nitrous and nitric forms produced was established.

Accordingly, nitrogen is rather well balanced (Figure 6). However, at the  $0.52 \text{ m}\cdot\text{d}^{-1}$  hydraulic rate, a slight difference between the amounts of the ammonia nitrogen removed and those of the oxidized forms produced was observed.

### Comparison of semi industrial and lab scale results

Table 5 compares laboratory (El Mossaoui *et al.*, 1999) and the current pilot scale results. It can be noticed that higher performances at  $0.52 \text{ m}\cdot\text{d}^{-1}$  were obtained on the semi industrial pilot ( $410 \text{ g N removed}\cdot\text{m}^{-3}\cdot\text{d}^{-1}$  instead of  $289 \text{ g N}\cdot\text{m}^{-3}\cdot\text{d}^{-1}$  obtained on the lab scale column). This could be explained by ventilation frequencies (6 ventilation periods instead of 3) and slightly higher temperatures on site.

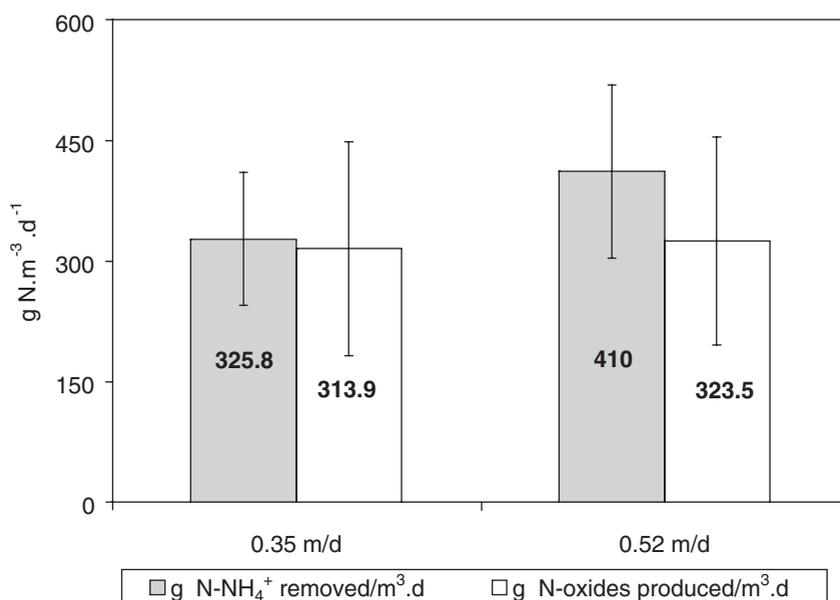
However, the very interesting volumetric nitrification rates obtained at lab scale are fairly well confirmed at semi industrial scale especially at  $0.35 \text{ m}\cdot\text{d}^{-1}$ .

### Other benefits: impact of the system on TOC and phosphate

During the lab scale study the leachate color became lighter after treatment. Total organic carbon (TOC) was measured at the inlet and the outlet of the device set up at the Montzen

**Table 5** Comparison of operating conditions and results obtained on lab scale columns and semi industrial scale pilot

	Hydraulic rate $\text{m}\cdot\text{d}^{-1}$	Batch (d <sup>-1</sup> )	(d <sup>-1</sup> )	T (°C)	$\text{NH}_4^+$ in $\text{mg N/l}$	$\text{NH}_4^+$ out $\text{mg/l}$	Efficiency $\text{g N removed}\cdot\text{m}^{-3}\cdot\text{d}^{-1}$
Lab scale	0.35	2	6	20–22	1,102	233	298.7
	0.52	3	3	20–22	1,102	608	289
Semi industrial	0.35	2	6	18–26	1,122.5	213.9	317.9
	0.52	3	6	20–25	1,086.9	284.7	410



**Figure 6** Nitrogen balances for the two hydraulic conditions (average and standard deviation)

site. TOC removal varied in the range of 11% to 47% and from 7% to 24% at the hydraulic rates of 0.35 m.d<sup>-1</sup> and 0.52 m.d<sup>-1</sup> respectively. In accordance with these results an elimination of about 90 gTOC.m<sup>-3</sup> of reactor.d<sup>-1</sup> at the rate of 0.35 m.d<sup>-1</sup> and 74 g at the rate of 0.52 m.d<sup>-1</sup> was calculated. The removal of the remaining organic matter could be achieved by biological and adsorption processes. This can lead to interesting savings on final treatment processes such as GAC (Granular Activated Carbon). It is well known that the duration of the aftercare period is a major issue of landfill management. This is the reason why various technologies are being tested to accelerate the stabilization process in the landfill. One of the ways to promote the mineralization in this bioreactor is to recirculate a nitrified effluent. In this case denitrification occurs in the landfill itself contributing to mineralization processes.

## Conclusions

Very high removal efficiencies (N, C and even refractory COD) can be obtained with MBR but the treatment costs are high. On the other hand, despite the high ammonia nitrogen inlet concentrations the infiltration-percolation reactor exhibits remarkable nitrification efficiency. Moreover, these performances are achieved without recirculation or dilution processes. The system can be classified as a low cost process. The current design of the system (hydraulic rate of 0.35 m.d<sup>-1</sup> divided into two feed periods, a surface area of 4.3 m<sup>2</sup>, and a ventilating system switched on for 6 × 15 min.d<sup>-1</sup>, consumes 0.47 kWh.d<sup>-1</sup> (0.18 kWh.d<sup>-1</sup> for the fan and 0.39 kWh.d<sup>-1</sup> for the pumps)), works out to a cost of 0.028 Euro/day (0.018 Euro/m<sup>3</sup> of treated effluent). By comparison, a pilot membrane bioreactor operating at this site on the same leachate consumed 14.4 kWh/m<sup>3</sup> of treated effluent (denitrification included). This simple system is thus very competitive and should be suitable for use in many of the situations that are encountered in the field when denitrification is not necessary. A preliminary mathematical model of this type of reactor has been proposed (Jupsin *et al.*, 2002). When properly operated such a type of low cost treatment could contribute efficiently to landfill leachate treatment and to the mineralization process in the deposits as well.

## References

- Bressi, G. and Favali, G. (1997). Use of a MBR (Membrane Bioreactor) in Leachate treatment. *Proceedings Sardinia 1997, Sixth International Waste Management and Landfill Symposium*, CISA Publisher, Cagliari, Italy, Vol. II, 261–274.
- El Mossaoui, M., Praet, E., Reuter, V. and Vassel, J.-L. (1999). Nitrification of landfill leachate using a modified infiltration-percolation process. *International Conference on Urban Pollution Technology*. The Hong Kong Polytechnic University. pp. 131–139.
- Imai, A., Iwami, N., Matsushige, K., Inamori, Y. and Sudo, R. (1993). Removal refractory organics and nitrogen from landfill leachate by the microorganisms attached activated carbon fluidized bed process. *Water Research*, 27(1), 143–145.
- Jupsin, H., Praet, E. and Vassel, J.-L. (2002). Low cost nitrification of leachate for better landfill management. *Wastecon 2002 International Waste Congress et Exhibition*, Durban South Africa. pp. 213–222.
- Jupsin, H., Praet, E. and Vassel, J.-L. (2003). About Refractory COD removal in MBR for leachate treatment, *SARDINIA 2003, Ninth International Waste Management and Landfill Symposium*, S. Margherita di Pula (Cagliari, Italy), 6–10 October 2003.
- Luning, L. and Notenboom, G. (1997). The membrane bioreactor, advanced leachate treatment. *Proceedings Sardinia 1997, Sixth International Waste Management and Landfill Symposium*, CISA publisher, Cagliari, Italy, Vol. II, 275–282.
- Praet, E., Reuter, V., Rouxhet, V. and Vassel, J.-L. (2000). Nitrification-dénitrification d'un lixiviat de CET par bioréacteur à membranes immergées, *Tribune de l'eau*, 53(603), 191–202.

- Praet, E., Jupsin, H., El Mossaoui, M., Rouxhet, V. and Vassel, J.-L. (2001). Use of a membrane bioreactor and an activated carbon adsorber for the treatment of a MSW landfill leachates. *Proc. 8th International Waste Management and Landfill Symposium*, Sardinia vol. II, pp. 351–360.
- Praet, E., Jupsin, H., Rouxhet, V. and Vassel, J.-L. (2002). Membrane Bioreactor (MBR) for old leachates treatment: from pilot plant to full-scale facility. *Second Asian – Pacific landfill symposium*. September 25–28, Seoul, Korea, pp. 102–108.
- Rees, J.F. (1982). Landfill management and leachate quality. *Effluent and Water Treatment Journal*, pp. 457–460.
- Robinson, H.D., Barber, C. and Maris, P.J. (1982). Generation and treatment of leachate from domestic wastes in landfills. *Water Pollution Control*, pp. 465–478.
- Vassel, J.-L., Vandevenne, L. and Berger, P. (1991) Epuration par lagunage aéré des lixiviats d'une décharge contrôlée de déchets ménagers. *Tribune de l'eau*, n. 553/5, pp. 7–18.
- Wens, P., De Langhe, P. and Staelens, B. (2001). Biological treatment of leachate by means of a biomembrane reactor. *Proceedings Sardinia 2001, Eighth International Waste Management and Landfill Symposium*, CISA Publisher, Cagliari, Italy, Vol. II, 169–178.