Control of methane emissions issuing from landfills: the Canadian case

JOSIANE NIKIEMA AND MICHÈLE HEITZ

Chemical Engineering Department, Faculty of Engineering, Université de Sherbrooke; 2500, Boulevard de l'Université, Sherbrooke, (Québec), J1K 2R1, Canada

ABSTRACT

During their storage in landfills, wastes are biodegraded, which results in the production of biogas and leachate. Over recent years, the handling of the leachate product has become one of major concern. However, in the case of biogas product, elimination or valorization processes are applied in a smaller proportion, even if the methane emissions, directly related to landfills, are some 25 % of the total anthropogenic methane emissions. Indeed, many older or smaller landfills are deprived of gas collection systems, thereby making impossible the application of gas combustion and/or valorization methods. Therefore, other processes have to be considered, *e.g.*, the biofiltration of methane. In this paper, the results of an experiment, undertaken to confirm the stability of the biofiltration system that has been developed at Université de Sherbrooke by the Biocom group, are presented. At a methane inlet concentration of around 7500 ppmv and a gas flow rate of $0.25 \text{ m}^3/\text{h}$, the conversion of the biofilter can be maintained at 22 % unchanged for a period of 150 days or more. Even after the cessation of methane feeding and biofilter irrigation for some 2 weeks, the biofilter performance was able to be restored, in only one week, to the same operating level as it was maintained before the deliberate shutdown.

1 INTRODUCTION

1.1 WASTES IN CANADA

During the last decade, it is to be noted that a continuous increase in the total amount of wastes generated in Canada took place. The total waste generated during the year 2004 was > 33 million metric tons (that is to say 790 metric tons per inhabitant, being 3 % higher than in 2002), of which 10 million metric tons originated from

residential sources. The remaining part arose from commercial and public institutions (50 %), and other sources. These wastes are mainly composed of organic materials (40 % m/m), papers (26 % m/m), plastics (9 %), glass, metals and others (Buchanan *et al.*, 2007).

Various reasons are advanced to explain this situation: the population increase, the increases in goods' consumption, caused in part by the higher levels of incomes, the changes in society that have resulted, for example, in increased need for non-reusable products, and the continuous evolution of technologies (Cameron *et al.*, 2005). Wastes can be substantially eliminated through recycling efforts (papers, plastics, metals and glass materials), composting and anaerobic digestion (organic wastes), thermal treatments (part of energy generation), and finally, by landfilling.

Increasingly, wastes are now being valorized through various recycling processes (e.g. 40-50 % of residential wastes may be recycled) but in Canada, the main way of disposing of wastes is still through landfilling, which affected, in 2005, around 75 % of the wastes arising during the same period. Indeed, about 10000 landfills (active and inactive, all of them requiring attention) presently exist in Canada (Nikiema *et al.*, 2007). The majority of them (83 %) are public institutions but private landfills also exist. Public landfills receive around 56 % of the wastes, private landfills receive the remainder. It must also to be mentioned that most landfills are now rather old. Indeed, some 30 % of landfills had, in 2002, a remaining useful life of less than 10 years. However, these landfills were still receiving more than half of the currently generated wastes (Cameron *et al.*, 2005).

1.2 The biogas and the leachate

During the degradation of wastes stored in landfills, a leachate and a biogas, both of which need to be handled, are generated. The quantities and compositions of these materials are influenced by various factors such as the types of wastes, their ages, etc. (Trebouet *et al.*, 2001).

The leachate is mainly composed of water, in which soluble and solid pollutant particules, are present, including minerals, e.g. iron, and organic matters (COD, up to 70000 mg/L; BOD, up to 56000 mg/L and TKN, up to 2000 mg/L) (Sanphoti *et al.*, 2006; Tränkler *et al.*, 2005; Visvanathan *et al.*, 2007). The leachate is generated during dehydratation of the biodegrading wastes or when rain water passes through the wastes; this explains its wide variations in composition. In Canada, in 2000, 46 % of municipal landfills were equipped with membranes, limiting the infiltration of external water into the site, while 18 % of landfills had collection systems installed for the leachate. These two kinds of landfills handled some 75 % of the total of landfilled wastes arising in the same year (Cameron *et al.*, 2005).

The other landfill product, the biogas, is a mixture of gases, composed principally of methane (30-70 % V/V), carbon dioxide (20-50 % V/V) (both greenhouse gases),

along with various sulphur compounds, volatile organic compounds, and others. Because of its high heating value during the landfill's early years (usually half that of natural gas), the biogas can be valorized, if collected, as an energy source (Nikiema *et al.*, 2007). In as much as is feasible, Canada Government policy encourages the energy recovery of biogas through combustion. There are some 50 landfills in Canada that collect their biogas and at least 30000 metric tons/day of methane are burned on each site. However, for the older or smaller landfills (i.e. < 200000 m³ of capacity), these are frequently deprived of gas collection systems, and thus valorization methods cannot be reasonably applied. Therefore, in order to avoid important methane emissions to atmosphere, biological processes, such as biofiltration, may be applied.

1.3 The methane biofiltration

Biofiltration is performed within a triphasic reactor, packed with stationary filter material, in which growth of the microorganisms is favoured (Delhoménie and Heitz, 2005). Indeed, methanotrophs are able to biodegrade the methane pollutant, and then generate, as in all biological processes, new biomass, salts, water and carbon dioxide, the latter product to a lesser extent than occurs in chemical oxidation processes, as presented in Equation 1.

$$CH_4 + xO_2 \rightarrow yCO_2 + zH_2O + Biomass + Salts$$
 (1)

Experiments conducted to date have confirmed that biofiltration is deemed suitable for the direct elimination of methane on landfill sites. The Biomet group, located at Université de Sherbrooke (Sherbrooke, Canada), has for more than 5 years, conducted research on the problematic of the methane issuing from landfills. The main interest was to determine the relationship between the measured biofilter performance and some operating parameters, including the concentrations of inlet methane and the nitrogen present in the nutrient solution. For example, during previous experiments, the nitrogen concentration, required for the proper operation of the biofilter, has been optimized and appears to be 0.75 g/L for a methane inlet concentration of between 7000 and 7500 ppmv (Nikiema *et al.*, 2005). The objective of this present study has been to confirm the stability of the biofiltration system, and its capacity to remain as efficient as it was initially, even after a substantial period of time (nearly one year).

2 MATERIALS AND METHODS

The lab-scale biofiltration system employed in this study is presented in Figure 1. It is mainly composed of 3 sections: 1) the polluted air generation section (humidified air

and pure methane (99 % V/V of purity); gas inlet temperature: ~ 20° C; inlet concentration of methane: 7500 ppmv; total gas flow rate: 0.25 m³/h); 2) the biofiltration system, which is composed of an up-flow biofilter (composed of two stages, each containing 33 cm of filtering material) and an irrigation system, and 3) the disposal of the exit gases and liquids.

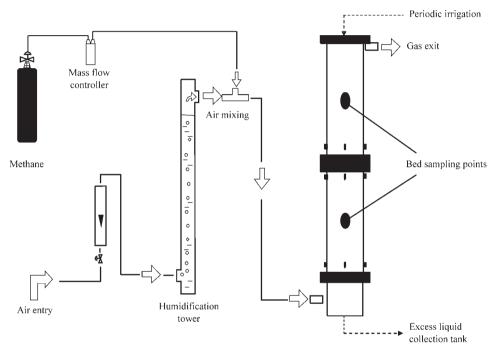


Figure 1. Methane biofiltration set-up.

It must be mentioned that the periodic bed irrigation (1 L/day for each biofilter) was performed using a nitrogen mineral salt solution, as described elsewhere (Delhoménie et al., 2007), containing 0.5 g/L of nitrogen. The filter bed used in the present study consists of an inorganic material having particles of around 5 cm mean diameter.

The following experimental results are expressed in terms of the inlet load, elimination capacity, carbon dioxide production and conversion, as described in Table 1.

Description	Calculation
Inlet load (g/m ³ .h)	$IL = \frac{C_{(CH_4)_{in}} \times Q}{V}$
Elimination capacity (g/m ³ .h)	$EC = \frac{\left[C_{(CH_{4}) \text{ in}} - C_{(CH_{4}) \text{ out}}\right] \times Q}{V}$
Carbon dioxide production (g/m ³ .h)	$\mathbf{P}_{\mathrm{CO}_{2}} = \frac{\left[\mathbf{C}_{(\mathrm{CO}_{2}) \text{ in }} - \mathbf{C}_{(\mathrm{CO}_{2}) \text{ out }}\right] \times \mathbf{Q}}{\mathbf{V}}$
Conversion (%)	$X = \frac{\left[C_{(CH_{4}) \text{ in }} - C_{(CH_{4}) \text{ out }}\right]}{C_{(CII_{4}) \text{ in }}} \times 100$

Table 1. Parameters used to express the results.

with: C: Concentration in g/m³ Q: Gas flow rate in m³/h V: Bed volume in m³.

3 RESULTS

Figure 2 presents the conversion (%) and inlet load (g/m³.h) in the inorganic-based biofilter, as a function of time (days). On day 0, the biofilter was inoculated with the lixiviate, being taken from another biofilter already treating methane (under similar operating conditions). At day 160, the biofiltration processing was stopped for a 2-week break period, during which the methane feed and biofilter irrigation ceased. However, the air feed was maintained for this period.

During this study, the inlet load remained at around 120 g/m³.h. It was noted that, after the 6 months of operation, the biofilter was still as efficient as it was at the start-up. After the imposed 2-week interruption period, the biofilter restart procedure (this time without inoculation) was achieved within a week, the biofilter reaching its maximum conversion rate soon after. In addition, no difference was detected in the performance of the biofilter, either before or after the imposed interruption (conversion = 22 %). During the whole of the experimental period, the 2 stages of the biofilter exhibited similar methane conversion. Indeed, around 48 % and 52 % of the methane elimination were achieved in stage 1 (lower) and stage 2 (upper) respectively.

Figure 3 presents data on the elimination capacity (g/m³.h) for, and the carbon dioxide production (g/m³.h) within, the biofilter, as a function of time (days). During the first six months of operation, the average elimination capacity was estimated to be \sim 30 g/m³.h, while the average carbon dioxide production was \sim 60 g/m³.h. However, after the planned interruption period, the average carbon dioxide production rate was

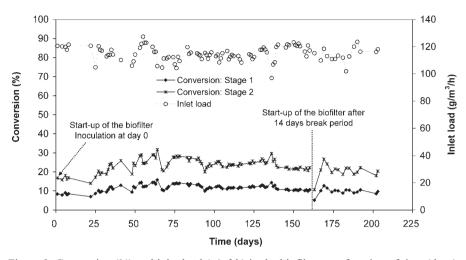


Figure 2. Conversion (%) and inlet load (g/m³.h) in the biofilter as a function of time (days).

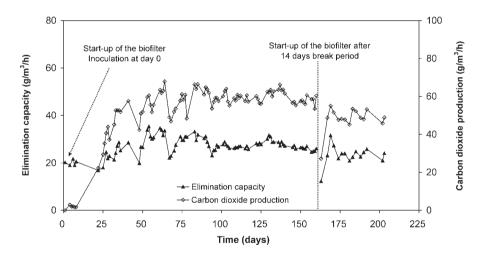


Figure 3. Elimination capacity (g/m³.h) and carbon dioxide production (g/m³.h) in the biofilter, as a function of time (days).

decreased, by ~ 15 %, indicating that the biomass growth rate was now at a higher value than that previously observed (Nikiema et al., 2005).

As reported for other methane treating biofilters, a good correlation can be observed between the methane elimination capacity and the carbon dioxide production, in the biofilter, except for the biofilter initial start-up, from day 0 to day 35. A possible explanation could be the multiplication rate of the microorganisms; it is at its highest

level at the start-up and then decreases with time until it becomes quite constant, when an equilibrium level is reached (consequently, CO_2 production keeps increasing until it reaches its maximum value, this being the phenomenon observed in the present biofilter). Another possible reason is the adsorption of methane on the biofilter's packing material. However, this possibility was later excluded because such phenomena would be reduced in intensity when the filter material is of an inorganic origin (Devinny and Ramesh, 2005). It is to be mentioned that higher methane conversions (compared to the average value of 22 % reported for this study) can be obtained, e.g. when higher nitrogen concentration is used (data not shown).

4 CONCLUSIONS

Wastes result principally from the expansion of the needs of humans living and their activities on the Earth. Thus, for many years in Canada, the total of human generated wastes has never stopped increasing. Along with this increase are created the problems associated with the environment maintenance and safety during the disposal of these wastes. In the case of methane emissions control, from older and/or smaller landfills, biofiltration could be a technically reliable solution. During the lab-scale experiments some of the parameters involved were identified as being important, some of them already having been optimized; e.g. the concentration of the input nitrogen. Therefore, the particular aim of this present study has been to confirm the stability of the biofiltration system undergoing development, i.e. its capacity to give the same performance, continuously, even after several operating months including a substantial process interruption period. Following the experimental operating period (6 months), the biofilter was observed to be still as efficient as it was at the test period commencement. In addition, the restarting of the biofilter, following the 2-week interruption period was readily performed. Indeed, the biofilter reached its maximum conversion performance after only one week. There was essentially no difference between the filter performance, before and after this deliberate interruption, except for the carbon dioxide production level which was at a slightly lower level in the post interruption operating period. Finally, as reported for other methane treating biofilter systems, it was observed that there was a good correlation between the methane elimination capacity and the carbon dioxide production.

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References

- Buchanan, B., Dewis, G., Lavergne, M., Mitchell, B. and Trépanier, H. (2007) Enquête sur l'industrie de la gestion et des administrations publiques des déchets secteurs des entreprises: 2004. In (Statistique Canada, Division des comptes et de la statistique de l'environnement), Système de comptabilité nationale, Ottawa, Ontario, pp. 48.
- Cameron, M., Elliott, A., Marshall, J. and Wang, J. (2005) L'activité humaine et l'environnement, Statistiques annuelles 2005; Article de fond: Les déchets solides au Canada. In (Statistique Canada, Division des comptes et de la statistique de l'environnement), Système de comptabilité nationale, Ottawa, Ontario, pp. 110.
- Delhoménie, M.-C. and Heitz, M. (2005) Biofiltration of air: A review. *Crit. Rev. Biotechnol.* 25(1-2): 53-72.
- Delhoménie, M.-C., Nikiema, J., Bibeau, L. and Heitz, M. (2007) A new method to determine the microbial kinetic parameters in biological air filters. *Chem. Eng. Sci.* submitted in 2007.
- Devinny, J.S. and Ramesh, J. (2005) A phenomenological review of biofilter models. *Chem. Eng.* J. 113(2-3): 187-196.
- Nikiema, J., Bibeau, L., Lavoie, J., Brzezinski, R., Vigneux, J. and Heitz, M. (2005) Biofiltration of methane: an experimental study. *Chem. Eng. J.* 113 (2-3): 111-117.
- Nikiema, J., Brzezinski, R. and Heitz, M. (2007) Elimination of methane generated from landfills by biofiltration: a review. *Rev. Environ. Sci. Bio/Technol.* DOI: 10.1007/s11157-006-9114-z.
- Sanphoti, N., Towprayoon, S., Chaiprasert, P. and Nopharatana, A. (2006) The effects of leachate recirculation, with supplemental water addition, on methane production and waste decomposition in a simulated tropical landfill. J. Env. Manage. 81(1): 27-35.
- Tränkler, J., Visvanathan, C., Kuruparan, P. and Tubtimthai, O. (2005) Influence of tropical seasonal variations on landfill leachate characteristics–Results from lysimeter studies. *Waste Manage*. 25(10): 1013-1020.
- Trebouet, D., Schlumpf, J.P., Jaouen, P. and Quemeneur, F. (2001) Stabilized landfill leachate treatment by combined physicochemical–nanofiltration processes. *Water Res.* 35(12): 2935-2942.
- Visvanathan, C., Choudhary, M.K., Montalbo, M.T. and Jegatheesan, V. (2007) Landfill leachate treatment using thermophilic membrane bioreactor. *Desalination* 204 (1-3): 8-16.