

Biofiltration of Methane Issued from Landfills, an Experimental Lab-Scale Study

*Authors: Nikiema J. and Heitz, M.**

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

* to whom all correspondence should be addressed
Tel.: 819-821-8000 ext. 62827
Fax: 819-821-7955
Email: Michele.Heitz@USherbrooke.ca

ABSTRACT

The aim of the study described in this paper was to examine the influence of methane inlet load on its biofiltration and thence, to optimize the quantity of nitrogen required for the maximum performance of the biofilter. Two filter materials were tested: a) an organic compost, and b) an inorganic filtration medium. The nitrogen was provided in the form of nitrates, through additions of a nutrient solution. These experiments, being undertaken with the use of an organic bed, have made it possible to put forward the poor performance of this type of material when utilized for methane elimination. It has also been established that the use of a nitrogen concentration of ~ 0.75 g/L leads to the largest elimination capacity at a methane inlet load of $75 \text{ g/m}^3/\text{h}$. In the inorganic bed, the particular influences of the methane inlet load, and of the nitrogen concentration, are more evident. Indeed, increases in the methane inlet load results in increases in the elimination capacity when the nitrogen substrate is not a reaction limiting factor. Optimum nitrogen concentrations, as a function of the inlet load, have also determined, thus for a methane inlet load of $95 \text{ g/m}^3/\text{h}$, the optimum nitrogen concentration is determined to be 0.75 g/L while for a methane inlet load of $55 \text{ g/m}^3/\text{h}$, it is only 0.5 g/L .

INTRODUCTION

Methane (CH_4) is among the most abundant of the naturally generated hydrocarbons. In Canada, the main sources of anthropogenic CH_4 emissions are; agriculture, landfills, natural gas delivery systems and petroleum exploitation activities. Presently, several solutions are being exploited in efforts leading to the control of this pollutant. Combustion methods, when applicable, and with or without heat recovery, are presently those most often used. In some particular cases, when methane gas valorization solutions cannot be economically applied, e.g., for older or smaller landfills, the biofiltration method has proven to be practical. This biological process exploits the ability of common micro-organisms, in particular the

methanotrophs, to degrade potential gaseous atmospheric pollutants, such as methane (Nikiema *et al.*, 2006).

For conducting this process in practice, compost-based beds are preferred because they already contain those natural nutrients that are essential for the growth of bacteria during biofiltration. Previously as reported in the literature various experiments, have been undertaken using composts as reactor filter materials (Hettiaratchi and Stein, 2001; Streese and Stegmann, 2003). Usually, this approach leads to good results being obtained for short period (a few months) experiments. Other experiments, conducted with inorganic materials (no intrinsic nutrients available) or soils-based beds, have also been reported in the literature (Park *et al.*, 2002; Nikiema *et al.*, 2005).

The objectives of our present research have been: 1) to optimize the quantity of nitrogen required by the micro-organisms employed, this compound being provided in the form of nitrates contained in a nutrient solution; and 2) to measure the influence of the CH₄ inlet load on its rate of biofiltration in the presence of an inorganic filter bed material.

MATERIALS AND METHODS

Figure 1 presents the flow sheet of the biofiltration system used for these experiments. The biofilter was an up-flow type system, made from 3 identical sections of 0.15 m internal diameter, and forming 3 reaction stages. The filter bed organic material tested in this study is a mature compost product, produced from vegetable residues and provided to us for these studies by GSI Environment inc. (Sherbrooke, Quebec, Canada). At the same time, an inorganic bed material was also tested.

During the study, each biofilter was humidified daily with nutrient solution at the rate of 0.1 L solution per L of filter bed. The monitoring of the biofilter performance (concentrations of methane (CH₄) and carbon dioxide (CO₂) at the entry and exit of each stage) was achieved via the use of 2 analyzers, the first, for total hydrocarbons, ex Horiba (Model FIA 510) and the second for CO₂, ex Siemens (Model Ultramat 22P). The average of the CH₄ inlet concentrations laid between 3.4 and 6.0 g/m³ (\pm 5%) while the total gas flow rate was 0.25 m³/h. The CH₄ used in our experiments (marketed by Praxair Inc. (Sherbrooke, Québec, Canada) had a purity of 99 % This pollutant gas was then mixed with ambient air (taken from compressed air network) and humidified to at least 90 % of saturation before use.

Two different nutrient solutions were tested during this study. Solution 1 was composed only of the nitrogen source, in the form of potassium nitrate dissolved in tap water. The nitrogen concentration of the solution was varied from 0.14 to 2 g/L, this Solution 1 being tested only in the organic filter bed. The composition of Solution 2 was similar to that previously employed by Fox *et al.* (1989). It contained nitrogen in the form of sodium nitrate (variable concentrations), phosphorus and micro-elements, dissolved in tap water. Solution 2 was tested on both filtering material types.

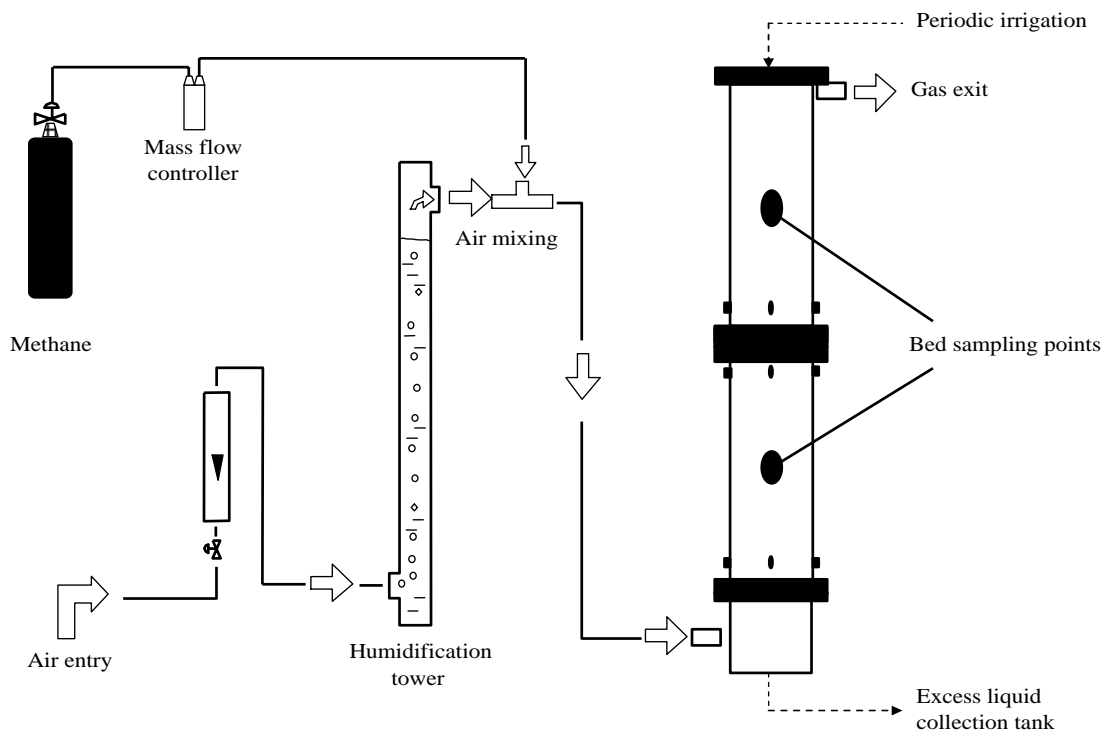


Figure 1: Methane biofiltration set-up

RESULTS AND DISCUSSION

Organic-based bed biofilter

Figure 2 presents the elimination capacity, expressed in $\text{g/m}^3/\text{h}$, being obtained in the compost-based bed, as a function of the nitrogen concentrations in Solution 1 and in Solution 2, being expressed in g/L . The inlet load was fixed at $75 \text{ g/m}^3/\text{h}$. In both nutrient solutions, the nitrogen concentrations were varied between 0.14 and 1. Additional nitrogen concentrations of 1.5 and 2 g/L , with Solution 1 and 0.003 g/L with Solution 2, were also tested.

With Solution 1, two domains are apparent: i.e., from 0.14 to 0.25 g-N/L , an increase of the EC with the nitrogen concentration: from 6 to 12 $\text{g/m}^3/\text{h}$ is noted; then, from 0.25 to 2 g-N/L , the EC is quite stable, remaining between 10 and 12 $\text{g/m}^3/\text{h}$. With Solution 2, the behaviour of the biofilter in terms of the EC is slightly different. First, between 0.003 and 0.14 g-N/L , the EC increases, from 7 to 13 $\text{g/m}^3/\text{h}$. Then, between 0.14 and 0.5 g-N/L , the EC is quite constant, remaining between 12 and 13 $\text{g/m}^3/\text{h}$. The highest EC for this set of experiments was 14 $\text{g/m}^3/\text{h}$, obtained at a nitrogen concentration of 0.75 g/L . When the N-concentration is increased, from 0.75 to 1 g/L , a 35% decrease of the EC is noted (9 versus 14 $\text{g/m}^3/\text{h}$). This later value is in the same range as those values obtained with Solution 1. Even when the nitrogen concentration of nutrient solution 2 is as low as 0.003 g/L , an EC of 7 $\text{g/m}^3/\text{h}$ is still obtained.

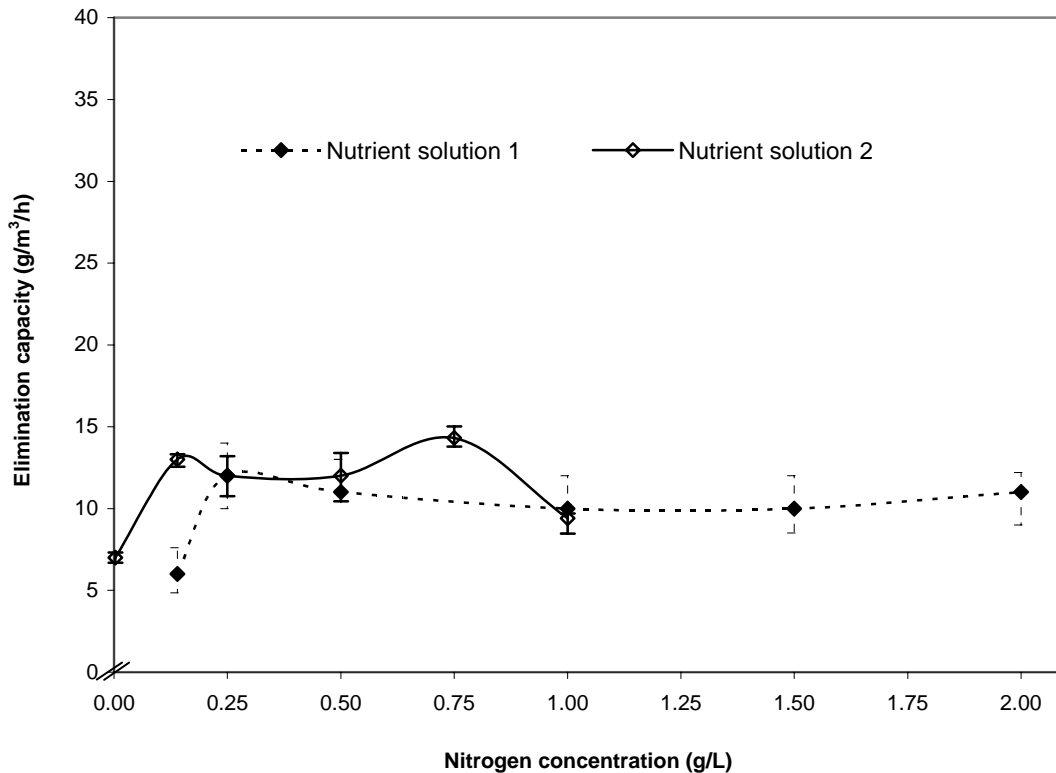


Figure 2: Elimination capacity, EC ($\text{g/m}^3/\text{h}$) for the compost-based bed as a function of the nitrogen concentration (g/L) for nutrient solutions 1 and 2: $\text{IL} = 75 \text{ g/m}^3/\text{h}$

During these experiments, the conversion remained below 20%. In addition, the nitrogen concentration seemed not to significantly affect the EC. Indeed, variations in the EC with the nitrogen concentrations were usually below 20 % (except with solution 1: from 0.14 to 0.25 g/L and with Solution 2: from 0.75 to 1 g/L), which is, statistically, not significant. Moreover, the presence of other nutrients in Solution 2, in addition to nitrogen, did not have a real positive influence on the CH_4 elimination in the compost-based biofilter. Indeed, during a previous experiment, it was found that the organic material was not always suitable for methane biofiltration since the EC was 2 times less than the one in the inorganic material (Nikiema *et al.*, 2005). The experiment, as presented here, shows that the nutrient solution is probably not the reason for this behaviour since the same low level of performance is provided by both solutions: 1 and 2.

Inorganic-based bed biofilter

Figure 3 presents the EC expressed in $\text{g/m}^3/\text{h}$, obtained with the inorganic-based bed, as a function of the nitrogen concentration (g/L), when Solution 2 was used. The inlet loads tested were 55 and 95 $\text{g/m}^3/\text{h}$. For each IL, it was therefore possible to determine the nitrogen concentration leading to the maximum EC. Indeed, at 55 $\text{g/m}^3/\text{h}$, the optimum nitrogen concentration is 0.5 g/L while for 95 $\text{g/m}^3/\text{h}$, it is 0.75 g/L . This confirms that the optimum nitrogen concentration varies, depending on the IL value. In addition, when the optimum

nitrogen concentration is exceeded, nitrogen becomes toxic to the micro-organisms by causing the reduction of the EC. Figure 3 also reveals that the increase of the IL, from 55 to 95 g/m³/h, causes the increase of the EC when the N-concentration is greater than 0.25 g/L. The explanation is that, for low nitrogen concentrations, a high IL is toxic for the micro-organisms, which is not the case when nutrients are available in sufficient quantities. During these experiments, the maximum conversions were about 48% and 40%, respectively for inlet loads of 55 g/m³/h and 95 g/m³/h.

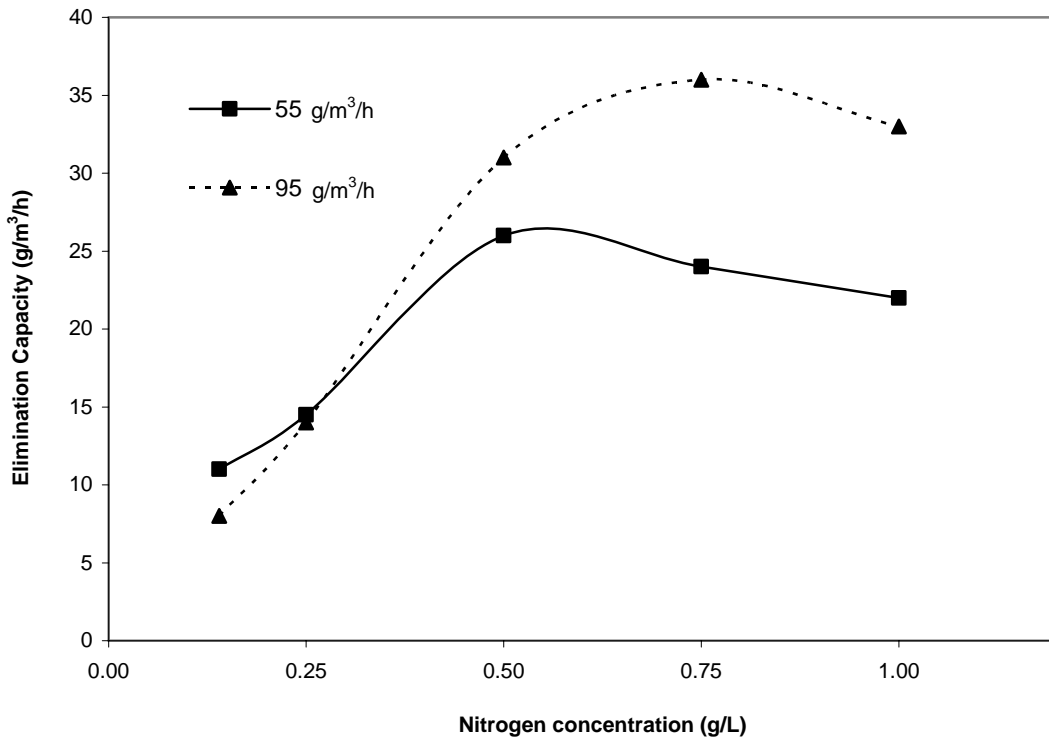


Figure 3: Elimination capacity (g/m³/h) in the inorganic-based bed as a function of the nitrogen concentration (g/L) for Solution 2: IL = 55 and 95 g/m³/h

Figure 4 presents the CO₂ production rate in the inorganic-based bed, expressed in g/m³/h, as a function of the nitrogen concentration (g/L) for Solution 2 (IL = 55 and 95 g/m³/h). It can be noted that both curves follow the same pattern which is approximately the same as that observed for the EC in Figure 3. This confirms that the CO₂ production rate is related to the EC in a biofilter.

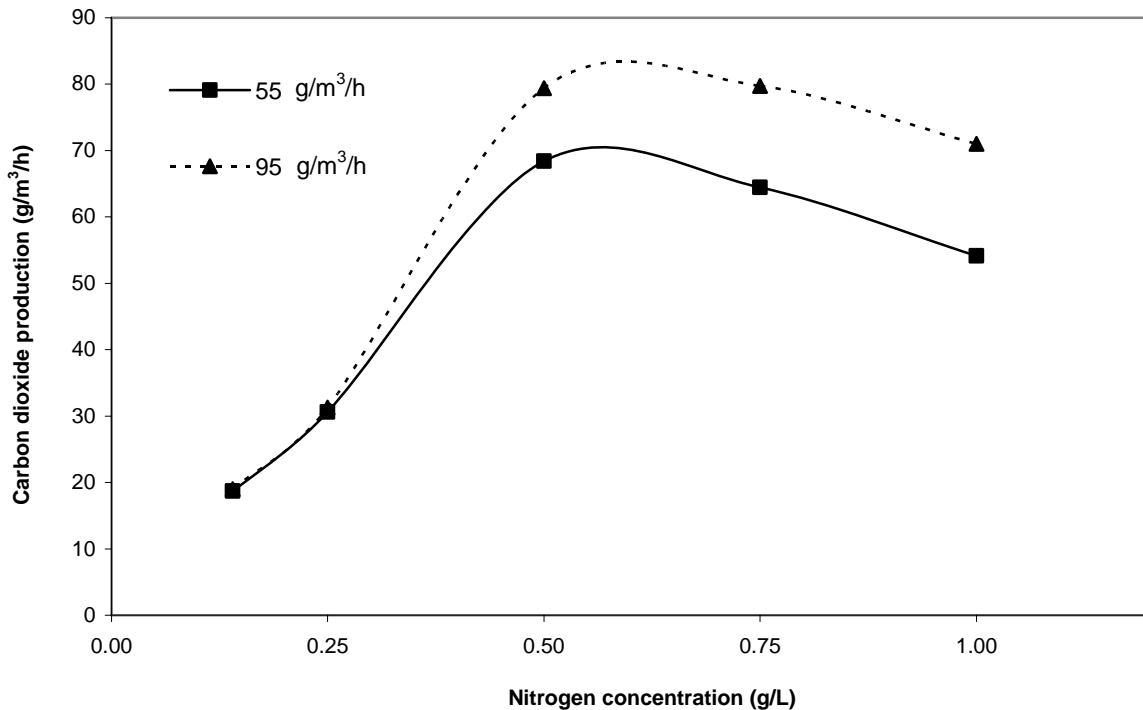


Figure 4: Carbon dioxide production rate ($\text{g/m}^3/\text{h}$) in the inorganic-based bed as a function of the nitrogen concentration (g/L) for Solution 2: $\text{IL} = 55$ and $95 \text{ g/m}^3/\text{h}$

CONCLUSION

The aim of this present study has been to determine the influence of the quantity of nitrogen nutrient compound, available during the methane biofiltration process, on the process. In the organic based-material bed, we found that the maximum value for the EC, i.e. $14 \text{ g/m}^3/\text{h}$, could be attained when the N-concentration was 0.75 g/L in a multi-component nutrient solution. However, the nitrogen level did not have great influence on the EC at concentrations above 0.25 g/L and the overall conversions always remained at values below 20 %. On the other hand, the inorganic based bed biofilter showed more classical trends, e.g., the nitrogen optimum concentrations were found to be 0.5 and 0.75 g/L , for inlet load values of 55 and $95 \text{ g/m}^3/\text{h}$ respectively. The carbon dioxide production rate in the inorganic biofilter showed trends which are approximately the same as those for the EC.

ACKNOWLEDGEMENTS

The authors are indebted to the Natural Science and Engineering Research Council of Canada (NSERC) for their financial support to this project. J. Nikiema would also like to thank NSERC for providing the scholarship for her doctoral studies support (Canada Graduate Scholarships Program). The authors also express their gratitude to Dr. Peter Lanigan for text revision.

REFERENCES

Fox A.B.G., Froland W.A., Dege J.E. and Lipscombs J.D. (1989) Methane monooxygenase from *Methylosinus trichosporium* OB3b: purification and properties of a three-component system with high specific activity from a type II methanotroph. *The American Society for Biochemistry and Molecular Biology*, 264 (17): 10023-10033.

Hettiaratchi J.P.A. and Stein V.B. (2001) Methanobiofilters (MBFs) and landfill cover systems for CH₄ emission mitigation. *Proceedings of the 17th International Conference on Solid Waste Technology and Management*, Philadelphia, Pa, Oct. 20-24, Published by the *Journal of Solid Waste Technology and Management*, Silver Spring, MD, USA, 465-476.

Nikiema J. and Heitz M. (2006) Elimination of methane generated from landfills by biofiltration: A review. *Re/Views in Environmental Science and Bio/Technology*, accepted in September 2006, 55 p.

Nikiema J., Bibeau L., Lavoie J., Brzezinski R., Vigneux J. and Heitz M. (2005) Biofiltration of Methane: an experimental study. *Chemical Engineering Journal*, 113(2-3): 111-117.

Park S., Brown K.W. and Thomas J.C. (2002) The effect of various environmental and design parameters on methane oxidation in a model biofilter. *Waste Management and Research*, 20(5): 434-444.

Streese J. and Stegmann (2003) Microbial oxidation of methane from old landfills in biofilters. *Waste Management*, 23: 573-580.

Key words: Biofiltration, methane, nitrogen, nitrates, compost, inorganic material.