

ANALYSIS OF THE EFFECTS OF TEMPERATURE, THE AMOUNT OF NUTRIENT SOLUTION AND THE CARBON DIOXIDE CONCENTRATION ON METHANE BIOFILTRATION

C. MENARD, A. AVALOS RAMIREZ, J. NIKIEMA & M. HEITZ

Department of Chemical Engineering and Biotechnological Engineering, Faculty of Engineering,
Université de Sherbrooke, Québec, Canada.

ABSTRACT

Landfill gas emissions contribute to the greenhouse effect due to the presence of methane (CH_4). CH_4 emissions from old and small landfills can be reduced by using biofiltration. The objective of this study was to optimize parameters that control CH_4 concentration in a biofilter. Temperature is one of the important parameters as well as the amount of nutrient solution (NS) supplied. The effects of the carbon dioxide (CO_2) concentration on CH_4 biofiltration were also studied. Four biofilters using an inorganic filter bed were studied under similar conditions: an inlet CH_4 concentration of 7000 ppmv and an air flow rate of 0.25 m³/h. A NS was supplied daily. The temperature was varied from 4°C to 43°C. The highest performance was obtained in the range of 31–34°C with an elimination capacity (EC) of 30 g CH_4 /m³/h for an inlet load (IL) of 80 g CH_4 /m³/h. The effect of the amount of NS supplied to the biofilter at ambient temperature was also analyzed. The EC was 23 g CH_4 /m³/h for both 101 L_{NS}/m³ v_{bed}/d and 34 L_{NS}/m³ v_{bed}/d, but it fell to 17 g CH_4 /m³/h at 17 L_{NS}/m³ v_{bed}/d. CO_2 concentrations were varied from 650 to 18,500 ppmv and no effect was noticed on the EC which remained constant at 18 g CH_4 /m³/h for an inlet load of 72 g CH_4 /m³/h.

Keywords: air treatment, biofiltration, carbon dioxide, environment, methane, nutrient solution, temperature.

1 INTRODUCTION

Atmospheric methane (CH_4) concentrations have increased from 715 ppbv during the industrial revolution (19th century) to 1785 ppbv in 2008 [1]. Up to 65% of emissions are due to anthropogenic sources such as fossil fuel combustion, agriculture, waste handling and rice cultivation [2]. CH_4 is the second most important greenhouse gas, just after carbon dioxide (CO_2). Its global warming potential (GWP) is 25 times higher than the one for CO_2 , based on a 100-year time horizon [3]. Among the anthropogenic sources, landfills contributed to 20% of the CH_4 emissions in Canada in 2007 [4], while the value was about 18% worldwide [5].

CH_4 recovery from landfills tends to be developed in Canada and 65 gas extraction systems were installed in 2007 [4]. Recent studies from nine landfill cells in France have shown that 92–97% of biogas can be recovered [6]. To achieve efficient energy valorization, CH_4 concentrations higher than 30–40% (v/v) are required with a minimum gas flow rate of 50 m³/h [7]. An alternative to energy valorization is flaring, which needs CH_4 concentrations higher than 20% (v/v) and a flow rate of 15 m³/h to be economically feasible [7]. When concentrations and flow rates are no longer appropriate for energy valorization or flaring, biofiltration is a bioprocess well adapted to control CH_4 emissions. It is generally the main control process for small and old landfills but it provides also a secondary treatment process for large and new landfills which have recovery installations [8].

Biofiltration is a triphasic biotechnology which uses microorganisms to eliminate pollutants like volatile organic compounds (VOC), volatile inorganic compounds (VIC) or greenhouse gases (GHGs) like CH_4 . The pollutant is transformed into water (H_2O), CO_2 , biomass and salts. The degradation process of CH_4 is divided into three steps. First, CH_4 is oxidized to methanol (CH_3OH) by an enzyme called methane monooxygenase (MMO). Methanol is then transformed

in formaldehyde (HCHO). This intermediary product is used to generate CO_2 , H_2O and biomass during the last step [9]. The bacteria responsible for CH_4 oxidation are called methanotrophic bacteria and are part of the methylotrophic bacteria able to assimilate C1 compounds (like CH_4 and methanol).

Engineered systems developed to optimize CH_4 biooxidation in landfills are either biocovers or biofilters. Biocovers are generally spread over an entire landfill area or a specific sector while biofilters are defined as fixed bed reactors, filled with a packing material and provided with a gas collection and drainage system [8].

Several variables need to be taken into account to control the microbial CH_4 oxidation such as the moisture content, temperature, oxygen availability, CH_4 concentration and addition of nutrients. Since microbial metabolism is limited by temperature, this parameter is one of the most important. Mesophilic cultures of methanotrophic bacteria have an optimal range from 20°C to 37°C to live and multiply [10]. Methanotrophic bacteria tolerant to cold have their optimum temperature under 20°C [11] and the microorganisms are still active down to $1-2^\circ\text{C}$ [12]. Laboratory batch experiments have shown an optimal temperature range of $30-36^\circ\text{C}$ and $25-35^\circ\text{C}$ in landfill soil biocovers [13, 14]. Results from field investigations indicated a higher CH_4 elimination for a temperature range from 9°C to 25°C than the range $2-9^\circ\text{C}$, 96% and 0–22%, respectively, using mechanically biologically treated waste as a biocover [15]. In western Canada, CH_4 conversion reached 33%, 55% and 89% in a landfill biocover, for respective minimal temperatures of 3°C (unheated bed), 8°C (heated bed) and 12°C (heated and temperature controlled bed) [11].

Similarly, the moisture content influences the CH_4 biooxidation rate as microorganisms require moisture to carry out their normal metabolic activity [16]. An optimal water level range should be sought for each filter material to prevent the drying-out of the filter bed or reversely, water clogging. The first event causes a significant reduction in the biodegradation rate while the second inhibits the transfer of oxygen and CH_4 and promotes the development of anaerobic zones [17, 18]. Several studies have dealt with the optimal range of moisture for CH_4 biofiltration using different filter beds. However, relatively few studies have analyzed the effect of the amount of nutrient solution (NS) supplied to the biofilter. According to our knowledge, one study was reported for toluene biofiltration, in a lab-scale fungal biofilter of 2.9 L, where the watering flow rate was decreased from $344 \text{ L}_{\text{NS}}/\text{m}^3 \text{ v}_{\text{bed}}/\text{d}$ to $34 \text{ L}_{\text{NS}}/\text{m}^3 \text{ v}_{\text{bed}}/\text{d}$ and also interrupted for 5 days [19]. The effect of nutrient addition is also important. In fact, some filter beds already contain the necessary macro and micronutrients to maintain an adequate microbial population [20]. However, an extra-addition of NS is needed in certain cases, particularly for inorganic filter bed [21].

The CO_2 concentration is not considered a key parameter for CH_4 biofiltration. However, high concentrations of CO_2 in the range from 30% to 65% (v/v) are often reported in landfill gas emissions [6, 8]. Previous studies report diverging results. A reduction of 16–30% in CH_4 uptake was observed in a forest soil continuously enriched with CO_2 at 200 ppmv above ambient levels [22]. In a different study, no significant effect in the rates of CH_4 oxidation was noticed for CO_2 concentrations ranging from 400 to 400,000 ppmv in laboratory experiments with landfill biocover [23]. However, the CO_2 respiration rates decreased with the high CO_2 concentrations.

The main objective of this study was to determine the optimal temperature range for CH_4 elimination with an inorganic filter bed at a CH_4 inlet concentration around 7000–7500 ppmv and a flow rate of $0.25 \text{ m}^3/\text{h}$. Two models to quantify the effect of the temperature were also tested. In addition, other experiments were performed to analyze the effect of the amount of NS supplied to the biofilter and the influence of CO_2 above the atmospheric concentration at ambient temperature (24°C).

2 MATERIALS AND METHODS

2.1 Experimental set-up

The experimental set-up is shown in Fig. 1. The upflow laboratory-scale biofilter column is made of Plexiglas with an internal diameter of 0.15 m. The biofilter is divided into three identical sections of 0.27 m high and was filled with an inorganic medium. Due to an existing confidentiality agreement, specific details about the characteristics of the filter bed are not available for publication at this time. The gas mixture is carried out at the bottom of the biofilter and consisted in mixing pre-humidified air and pure CH₄ (Praxair Inc., Québec, Canada) in the desired concentration. The effluent gas is sent to an evacuation system.

2.2 Operating conditions

Experiments were carried out on two biofilters to evaluate the influence of the temperature. Both were operated under the same inlet air flow rate of 0.25 m³/h at a CH₄ concentration around 7000–7500 ppmv. The initial temperature was fixed at 24°C. After 20 days of operation, one biofilter (BFH) was covered with both a silicon heating unit and an aluminum thermal blanket to increase the temperature from 24°C to 43°C. The second biofilter (BFL) was put into a temperature controlled chamber to decrease the temperature. Temperatures tested for BFH were 25°C, 31°C, 34°C, 41°C and 43°C, and 25°C, 14°C and 4°C for BFL. The amount of NS was constant at 67 L_{NS}/m³_{V bed}/d.

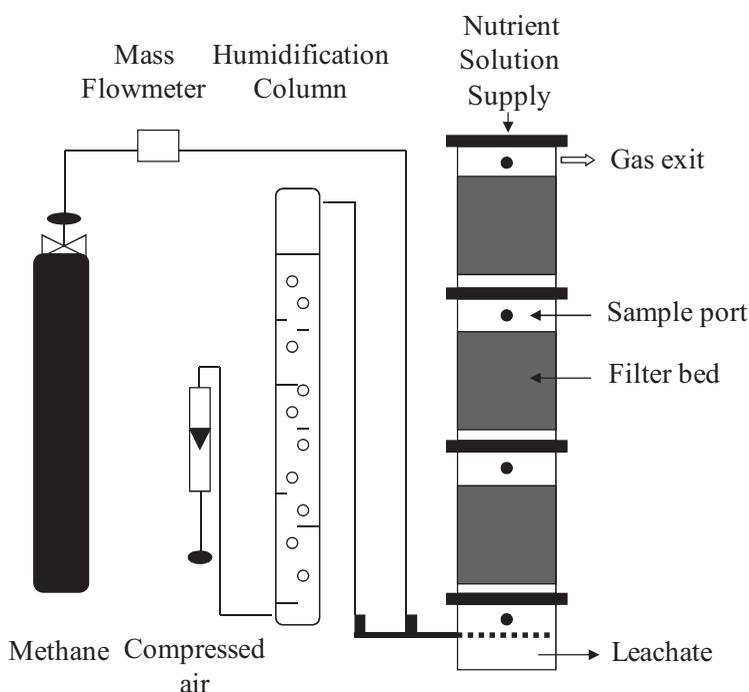


Figure 1: Laboratory-scale biofiltration system.

A third biofilter (BFA) was used to evaluate the influence of the amount of NS supplied with an inlet air flow rate of 0.25 m³/h and at a CH₄ concentration around 7000–7500 ppmv at ambient temperature (24°C). The amount of NS supplied ranged from 17 L_{NS}/m³ V_{bed}/d to 101 L_{NS}/m³ V_{bed}/d.

CO₂ concentrations were varied in a fourth biofilter (BFC) from 650 to 18,500 ppmv. BFC was operated at ambient temperature (24°C) with an amount of NS of 67 L_{NS}/m³ V_{bed}/d. The detailed composition of the NS for macro and micronutrients is described in Table 1.

2.3 Parameters for analyzing biofilter performance

The performance of a biofilter is expressed in terms of the inlet load (IL) (g CH₄/m³/h), the elimination capacity (EC) (g CH₄/m³/h), the conversion rate X (non-dimensional) and the CO₂ production rate PCO₂ (g CO₂/m³/h) as shown below:

$$IL = \frac{C_{in} * Q}{V_{bed}} \quad (1)$$

$$EC = \frac{(C_{in} - C_{out}) * Q}{V_{bed}} \quad (2)$$

$$X = \frac{C_{in} - C_{out}}{C_{in}} \quad (3)$$

$$PCO_2 = \frac{(C_{CO_2,out} - C_{CO_2,in}) * Q}{V_{bed}} \quad (4)$$

where Q is the total air flow rate (m³/h), V_{bed} is the packing bed volume (m³), C_{in} is the CH₄ inlet concentration (g/m³), C_{out} is the CH₄ outlet concentration (g/m³), $C_{CO_2,in}$ is the CO₂ inlet concentration (g/m³/h) and $C_{CO_2,out}$ is the CO₂ outlet concentration (g/m³/h).

The inlet and outlet CH₄ concentrations present in the gas phase were measured by means of a FIA-510 total hydrocarbon analyzer (Horiba, USA). CO₂ concentrations were analyzed with a

Table 1: Composition and concentration of the macro and micronutrient solution.

Macronutrients	Concentration (mg/L)	Micronutrients	Concentration (µg/L)
NaNO ₃	3038	ZnSO ₄ , 7H ₂ O	576
K ₂ SO ₄	170	MnSO ₄ , 7H ₂ O	466
MgSO ₄ , 7H ₂ O	37	H ₃ BO ₃	124
CaCl ₂ , 2H ₂ O	7	NaMoO ₄ , 2H ₂ O	96
KH ₂ PO ₄	530	CoCl ₂ , 6H ₂ O	96
Na ₂ HPO ₄	860	KI	166
		CuSO ₄ , 5H ₂ O	250
		FeSO ₄ , 7H ₂ O	112

portable gas analyzer system (Ultramat 22P, Siemens AG, Germany). The bed pressure drop was measured with a differential manometer (Type 4, Air Flow Developments Ltd., UK). The temperature inside the filter bed was monitored with T-type thermocouples (18G, Omega, USA). The moisture content of the filter bed was calculated by subtracting the mass of a sample of the filter bed before and after drying in an oven at 105°C and divided by the mass of the sample.

2.4 Parameters for modeling the effect of the temperature

Several models have been used to quantify the effect of the temperature on the growth of different microorganisms like Arrhenius or Esener. These models are based originally on the determination of microkinetic parameters such as the maximum specific growth rate (μ_{\max}) by batch experiments [24, 25].

To perform kinetic analysis in a biofilter, another approach has been considered, the macrokinetic approach, which is related to the pollutant biodegradation rate [26]. In this case, the kinetic parameters are defined with models based on the Michaelis–Menten approach. It has been shown that macrokinetic models fit well to the experimental EC [26–28].

In the present study, we used the macrokinetic approach in both the modified model of Arrhenius [29] and the model of Esener [30].

The modified Arrhenius equation is given below:

$$EC = A \cdot e^{(-E_a/RT)} \quad (5)$$

where A is a pre-exponential factor ($\text{g}/\text{m}^3/\text{h}$), E_a is the activation energy for CH_4 biodegradation (kJ/mol), R is the universal gas constant ($\text{kJ}/\text{mol}/\text{K}$) and T is the absolute temperature (K).

The second model is the modified Esener model [30]:

$$EC = \frac{A' \cdot e^{(-E_1/RT)}}{1 + k \cdot e^{(-E_2/RT)}} \quad (6)$$

where A' ($\text{g}/\text{m}^3/\text{h}$) and k (dimensionless) are both pre-exponential factors and E_1 , E_2 are the activation energy for CH_4 biodegradation and for the thermal denaturation processes (kJ/mol), respectively. The optimum temperature (T_{opt}) was calculated thanks to the following equation obtained by setting the first derivative of eqn (6) with respect to T equal to zero:

$$T_{opt} = \frac{E_2}{\left[R \cdot \ln \left(k \cdot \left(\frac{E_2}{E_1} - 1 \right) \right) \right]} \quad (7)$$

3 RESULTS AND DISCUSSION

3.1 Temperature experiments

3.1.1 Experimental results

For the two biofilters BFH and BFL, the IL, the PCO_2 , the EC and temperature (T) as a function of time are presented in Figs 2 and 3. The IL was nearly constant during all the experiments with a mean value of $80 \pm 5 \text{ g CH}_4/\text{m}^3/\text{h}$ for the two biofilters.

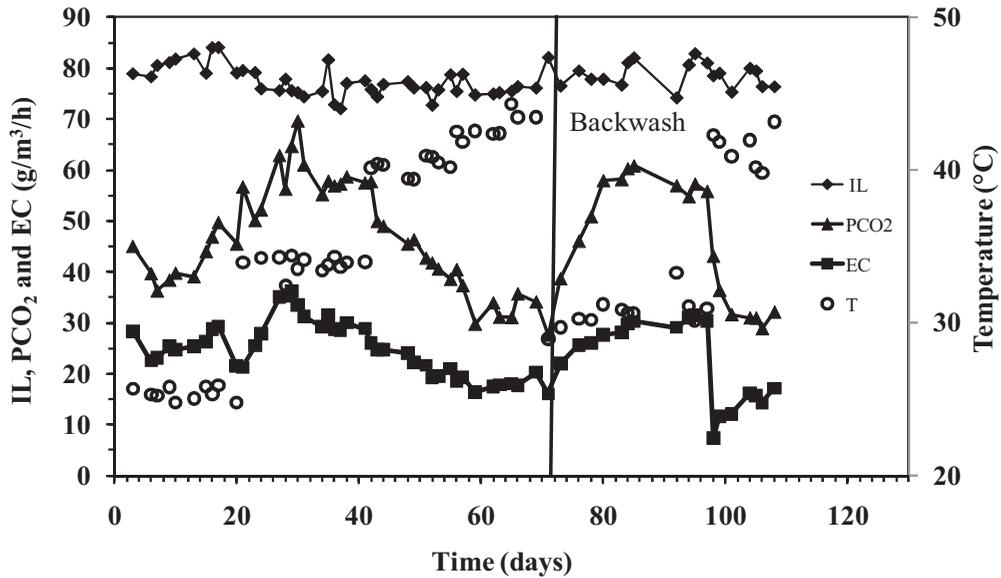


Figure 2: Inlet load (IL), carbon dioxide production rate (PCO₂), elimination capacity (EC) and temperature (T) as a function of time for BFH.

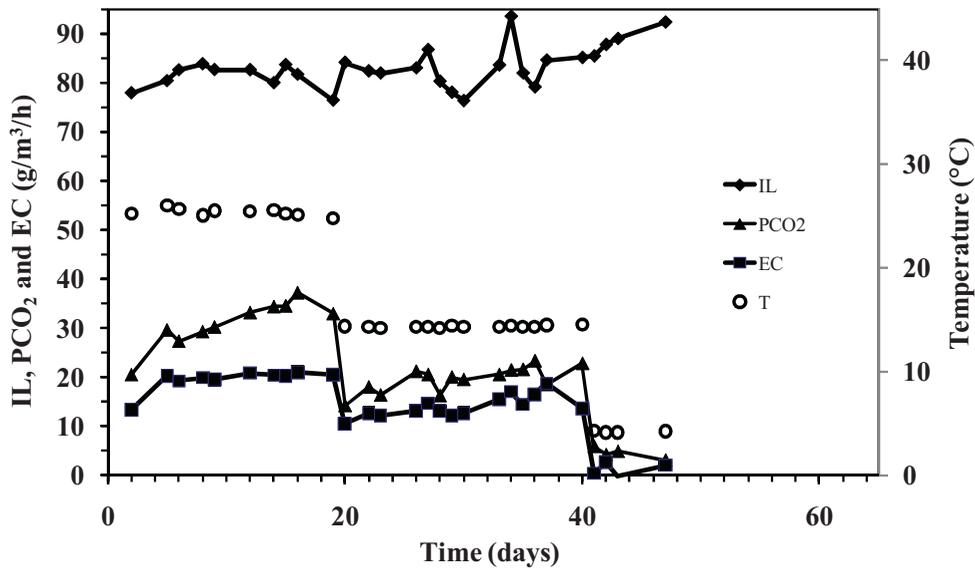


Figure 3: Inlet load (IL), carbon dioxide production rate (PCO₂), elimination capacity (EC) and temperature (T) as a function of time for BFL.

Both BFH and BFL were operated under the ambient room temperature of 24°C during the first 20-day period. The average measured temperature inside the filter bed was 25.4°C. From day 20 to 40, the average temperatures were 33.6°C and 14.3°C for BFH and BFL, respectively. BFH was operated for 15 days at 40.3°C and 14 days at 42.9°C, while BFL was operated one more week at 4.2°C. To confirm the optimal range of temperature, BFH was operated for 27 days more at 31.1°C after a backwash of the biofilter. The temperature was then increased to 41.5°C during 10 days.

At ambient temperature (24°C), the EC averaged 23 ± 2 g CH₄/m³/h for BFH and BFL. At 33.6°C, the EC reached a plateau of 30 ± 1 g CH₄/m³/h. For temperatures higher than 34°C, the EC decreased with temperature. At 40.3°C, the EC varied from 26 to 21 g CH₄/m³/h and it remained constant at 18 ± 1 g CH₄/m³/h for 14 days at 42.9°C. After the backwash of the biofilter, the EC reached a value similar to the one at 33.6°C. These results confirmed that the optimal temperature range was from 31°C to 34°C. Increasing the temperature by steps of 10°C resulted in a decrease of biofilter performance with a drop in EC to 7 g CH₄/m³/h at 41.5°C and then a stabilization at 14 ± 4 g CH₄/m³/h which resembled the previous results at 42.9°C.

The decrease in temperature had a negative effect on BFL's performance. The first temperature drop from 24°C to 14.3°C resulted in a decrease of the EC from 23 ± 2 to 14 ± 4 g CH₄/m³/h. From 14.3°C to 4.2°C, BFL showed nearly a complete inhibition of CH₄ oxidation with an EC of 1.2 ± 1.4 g CH₄/m³/h.

The CO₂ production rate followed the EC trend with high values up to 58 ± 5 g CO₂/m³/h at 33.6°C and low ones of 4 ± 1 g CO₂/m³/h at 4.2°C. The decrease in temperature by steps of 10°C implied a drastic change of operating conditions. The degrading bacteria were still able to remove CH₄ at 14.3°C with a conversion of $17 \pm 2\%$ but when the temperature dropped to 4°C, conversion was as low as $1 \pm 2\%$. Complete inhibition at low temperatures has already been reported by several authors [13, 31]. Reversely, no inhibition was noticed when the temperature was increased with a conversion of $30 \pm 3\%$ and $24 \pm 1\%$ for 40.3°C and 42.9°C, respectively. Temperatures higher than 45°C were not studied due to reasons of laboratory safety. Furthermore, this temperature has already been shown to inhibit CH₄ oxidation [13, 32].

This response to temperature results from the type of microorganisms responsible for CH₄ degradation. As it was shown in a previous study, *Methylocystis parvus* appeared to be the dominant CH₄-degrading bacteria in a biofilter used to treat CH₄ [21]. This bacteria is mesophilic with an optimal range of temperature for growth between 23–25°C and 31–34°C [33].

The values of pressure drop (ΔP) for BFH and BFL are presented in Table 2. The initial ΔP was similar for the two biofilters at 0.04 cm H₂O/m. At day 47, BFL had a final ΔP of 0.07 cm H₂O/m

Table 2: Pressure drop of BFL and BFH.

Biofilter	Period (days)	Temperature (°C)	Total ΔP (cm H ₂ O/m)
BFH	1 – 20	25.4	0.04
	21 – 41	33.6	0.04 – 0.05
	42 – 55	40.3	0.05 – 0.08
	56 – 69	42.9	0.09 – 0.11
	(backwash)	70 – 97	31.1
BFL	98 – 108	41.5	0.5 – 0.7
	1 – 19	25.4	0.04
	20 – 40	14.3	0.04 – 0.06
	41 – 47	4.2	0.07

and at day 69, BFH's ΔP was 0.11 cm H₂O/m. These values of ΔP are low but BFH was backwashed on day 69 because of a high amount of biomass visible in the biofilter. After the backwash, the values of ΔP are higher because of water accumulation in the biofilter. The backwash procedure used in this study consisted of pouring tap water (~4 L) in each of the three biofilter sections individually. However, the 10°C temperature increase at day 98 may have killed a significant amount of biomass by thermal denaturation, therefore increasing BFH's ΔP from 0.5 cm H₂O/m to 0.7 cm H₂O/m.

3.1.2 Quantification of the effect of temperature

Figure 4 presents the experimental data of the EC as a function of temperature and the models. The coefficients for the Arrhenius type model and the Esener type model are given in Table 3. The determination coefficients obtained for the Arrhenius (14–34°C) and Esener (4–41°C) models fitted to experimental data were 0.972 and 0.975, respectively. From 14°C to 34°C, the EC followed an exponential trend as

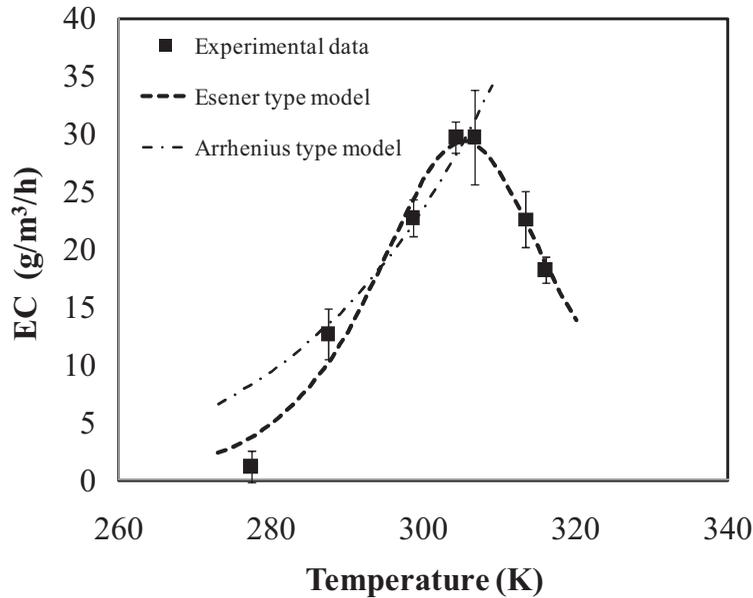


Figure 4: Effect of temperature on elimination capacity (EC) on methane in an inorganic biofilter.

Table 3: Temperature coefficients for Arrhenius and Esener models.

Coefficients	
A, g/m ³ /h	8.5E + 6
E _a , kJ/mol	32
A', g/m ³ /h	1.6E + 13
k (-)	1.7E + 25
E ₁ , kJ/mol	67
E ₂ , kJ/mol	148

predicted by the Arrhenius equation. However, the EC decreased after reaching the T_{opt} in the biofilter. The T_{opt} calculated from eqn (7) was 32°C. This value lies within a typical range for mesophilic microorganisms [10, 13, 14] and it was in the optimal range of temperature observed experimentally from 31°C to 34°C.

The results of the Esener type model are significant according to a t -test with a t value of 14 in a confidence interval of 95% ($n = 7$), as are the results of the Arrhenius type model with a t of 13 in the same confidence interval ($n = 4$).

The activation energy is generally underestimated with the use of the Arrhenius type model [25] which is consistent with the values presented in Table 3 where $E_a = 32 \text{ kJ/mol} < E_1 = 67 \text{ kJ/mol}$. The Esener model introduces a term which predicts the decrease of the macrokinetic parameter after T_{opt} . In fact, E_2 is defined as the activation energy for thermal denaturation and is generally higher than the activation energy for growth [34]. In our case, E_2 is two times higher than E_1 . For comparison, Gebert *et al.* have calculated an energy activation of 74.5 kJ/mol for CH_4 oxidation in a biofilter for a temperature rise from 10°C to 20°C [17].

3.2 Experiment on the amount of nutrient solution

Figure 5 presents the EC and the amount of NS for an IL of $80 \pm 2 \text{ g CH}_4/\text{m}^3/\text{h}$. A period of adaptation was noticed from day 1 to day 14 with a maximum EC of $29 \pm 1 \text{ g CH}_4/\text{m}^3/\text{h}$. Decreasing the amount of NS from $101 \text{ L}_{\text{NS}}/\text{m}^3 \text{ v}_{\text{bed}}/\text{d}$ to $34 \text{ L}_{\text{NS}}/\text{m}^3 \text{ v}_{\text{bed}}/\text{d}$ had no visible effect on biofilter performance. The EC remained constant around $23 \pm 1 \text{ g CH}_4/\text{m}^3/\text{h}$. Reducing the amount of NS to $17 \text{ L}_{\text{NS}}/\text{m}^3 \text{ v}_{\text{bed}}/\text{d}$ induced a decrease of the EC to $17 \pm 3 \text{ g CH}_4/\text{m}^3/\text{h}$.

No important increase in the P was noticed when the amount of NS was decreased from $101 \text{ L}_{\text{NS}}/\text{m}^3 \text{ v}_{\text{bed}}/\text{d}$ to $34 \text{ L}_{\text{NS}}/\text{m}^3 \text{ v}_{\text{bed}}/\text{d}$ with 0.09 cm $\text{H}_2\text{O}/\text{m}$ and 0.14 cm $\text{H}_2\text{O}/\text{m}$ at the end of each period, respectively. After the backwash of the biofilter on day 56, the P at $17 \text{ L}_{\text{NS}}/\text{m}^3 \text{ v}_{\text{bed}}/\text{d}$ has decreased to 0.04 cm $\text{H}_2\text{O}/\text{m}$.

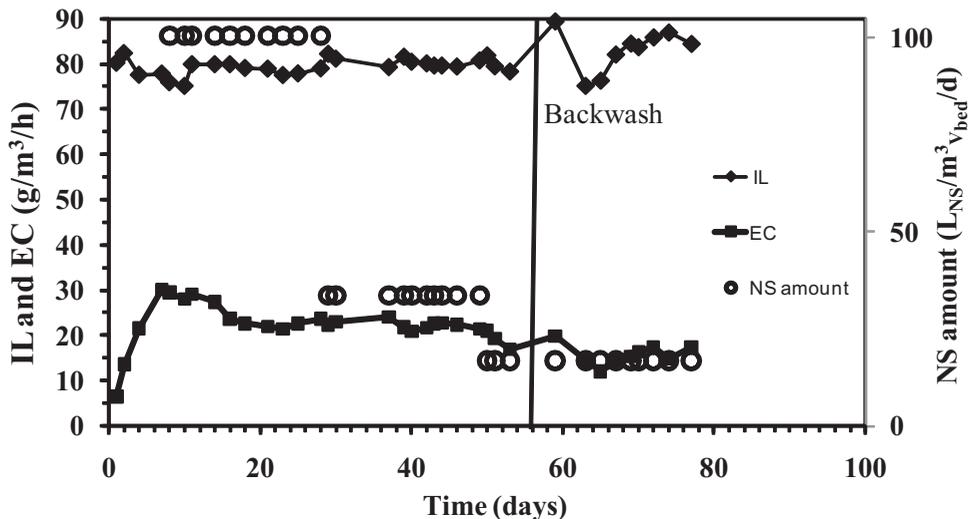


Figure 5: Inlet load (IL), elimination capacity (EC) and nutrient solution (NS) amount as a function of time for BFA.

The moisture content of each biofilter section was measured at the three NS flow rates. While it averaged 36% (water/filter bed sample, w/w) for the three sections for both $101 \text{ L}_{\text{NS}}/\text{m}^3 \text{ v}_{\text{bed}}/\text{d}$ and $34 \text{ L}_{\text{NS}}/\text{m}^3 \text{ v}_{\text{bed}}/\text{d}$, it decreased to 27% at $17 \text{ L}_{\text{NS}}/\text{m}^3 \text{ v}_{\text{bed}}/\text{d}$.

A NS is necessary due to the use of an inorganic filter bed. No water accumulation was observed in the top section of the biofilter which could explain the low ΔP . On the other hand, the CH_4 conversion decreased by 33% in the bottom section when the amount of NS was reduced from $34 \text{ L}_{\text{NS}}/\text{m}^3 \text{ v}_{\text{bed}}/\text{d}$ to $17 \text{ L}_{\text{NS}}/\text{m}^3 \text{ v}_{\text{bed}}/\text{d}$. No drying-out of the filter bed was observed. However, it may be hypothesized that the nutrient uptake is higher in the top section and therefore deprived the bottom section from the micro and macronutrients. Considering these details, a minimal amount of NS of $34 \text{ L}_{\text{NS}}/\text{m}^3 \text{ v}_{\text{bed}}/\text{d}$ is required as there will be an optimal distribution of the NS and no water accumulation along the biofilter.

3.3 Experiment on the concentration of carbon dioxide

Figure 6 presents the IL, PCO_2 , EC and CO_2 concentration as a function of time for BFC. The IL was fixed at $73 \pm 2 \text{ g CH}_4/\text{m}^3/\text{h}$. The biofilter was operated during 99 days and no variation of the EC was noticed with $18 \pm 1 \text{ g CH}_4/\text{m}^3/\text{h}$ for CO_2 concentrations varying from 650 to 18,500 ppmv. These results are coherent with previous experiments in the literature where no effect was visible in CH_4 oxidation rates for CO_2 concentrations varying from 400 to 400,000 ppmv in landfill cover soils [23]. The results are different from the experiments led on forest soil where the CH_4 consumption rate was reduced by 30% for a CO_2 concentration 200 ppmv higher than the atmospheric concentration (400 ppmv) [22].

CO_2 concentrations in forest soils are generally lower (0.04%, v/v) than in landfill soils (30–65%, v/v) [6, 8, 22]. It may be hypothesized that methanotrophs from forest soils would be more affected by variations in CO_2 concentrations than the methanotrophs responsible for CH_4 degradation in landfill soils.

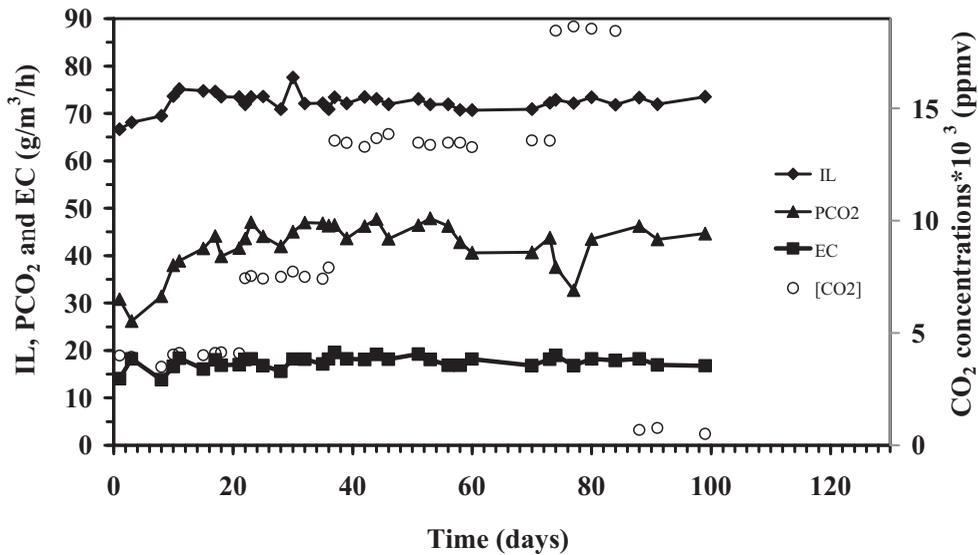


Figure 6: Inlet load (IL), elimination capacity (EC), carbon dioxide production rate (PCO_2) and CO_2 concentrations as a function of time for BFC.

4 CONCLUSION

The main objective of this study was to evaluate the effect of the temperature on CH₄ removal by biofiltration. Eight temperatures were tested from 4°C to 43°C. The effect of the temperature was well quantified, thanks to the modified Esener model. From this model, the optimum temperature calculated was 32°C which was in the range observed experimentally. At this temperature, the highest EC was observed with an average of 30 ± 1 g CH₄/m³/h for an IL of 80 ± 5 g CH₄/m³/h. The decrease of the amount of NS from $101 \text{ L}_{\text{NS}}/\text{m}^3 \text{ v}_{\text{bed}}/\text{d}$ to $34 \text{ L}_{\text{NS}}/\text{m}^3 \text{ v}_{\text{bed}}/\text{d}$ appeared to have no effect on the EC. However, at $17 \text{ L}_{\text{NS}}/\text{m}^3 \text{ v}_{\text{bed}}/\text{d}$, a decrease of the EC was observed at 17 ± 3 g CH₄/m³/h. A minimal amount of NS of $34 \text{ L}_{\text{NS}}/\text{m}^3 \text{ v}_{\text{bed}}/\text{d}$ should be added to the biofilter to ensure an equal distribution of the NS. The study regarding CO₂ concentrations varying from 650 to 18,500 ppmv showed no effect on the EC which remained constant at 18 ± 1 g CH₄/m³/h.

The results obtained in this study highlight the importance of both the temperature and the amount of NS for an inorganic based-bed biofilter and may have to be taken into consideration in landfill management. Landfills are open-space operated and undergo high temperature variations annually. Because of the decrease in microorganism activity under 14°C, it is essential to design the biofilter adequately in anticipation of the cold season to maintain a minimal temperature in filter bed and ensure the microbial degradation of CH₄. Although the objective would be to remain at the optimum temperature, it should be noted that microbial activity could be sustained even during the cold season. Furthermore, an appropriate way to spray the NS has to be installed to promote a good homogeneity of nutrient supply in the filter bed and also to control the supply flow rate to minimize either the drying-out or water clogging phenomena. Finally, the range of CO₂ concentrations tested did not show any inhibition of CH₄ oxidation. However, higher concentrations of CO₂ should be tested at a pilot scale to confirm this trend for real situations.

ACKNOWLEDGEMENTS

This work was supported by a Strategic grant from the Natural Sciences and Engineering Council of Canada to Michèle Heitz in partnership with le Centre de Recherche Industrielle du Québec and Viaporc Inc. and by a Discovery grant from the Natural Sciences and Engineering Council of Canada to Michèle Heitz. The authors are also thankful to Matthieu Girard, PhD student in chemical engineering and biotechnological engineering, for English text revision.

REFERENCES

- [1] Leigh Mascarelli, A., A sleeping giant? *Nature*, **3**, pp. 46–49, 2009.
- [2] Olivier, J.J.M. & Berdowski, J.G.J., *Global Emission Sources and Sinks, The Climate System*, A.A. Balkema Publishers, 2001.
- [3] Solomon, S., Qin, D., Manning, M., Alley, R.B., Berntsen, T., Bindoff, N.L., Chen, Z., Chidthaisong, A., Gregory, J.M., Hegerl, G.C., Heimann, M., Hewitson, B., Hoskins, B.J., Joos, F., Jouzel, J., Kattsov, V., Lohmann, U., Matsuno, T., Molina, M., Nicholls, N., Overpeck, J., Raga, G., Ramaswamy, V., Ren, J., Rusticucci, M., Somerville, R., Stocker, T.F., Whetton, P., Wood, R.A. & Wratt, D., *Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, Cambridge University Press: Cambridge, United Kingdom and New York, NY, USA, 2007.
- [4] Environnement Canada, Sources et puits de gaz à effet de serre au Canada. Rapport d'inventaire national 1990–2007. Sources et puits de gaz à effet de serre au Canada. 978-1-100-12999-0, pp. 1–747, 2009.

- [5] Bogner, J., Abdelrafie Ahmed, M., Diaz, C., Faaij, A., Gao, Q., Hashimoto, S., Mareckova, K., Pipatti, R. & Zhang, T., *Waste Management, In Climate Change 2007: Mitigation. Contribution of Working Group III to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, Chapter 10, Cambridge University Press: Cambridge, United Kingdom and New York, NY, USA, pp. 585–618, 2007.
- [6] Spokas, K., Bogner, J., Chanton, J.P., Morcet, M., Aran, C., Graff, C., Golvan, Y.M. & Hebe, I., Methane mass balance at three landfill sites: what is the efficiency of capture by gas collection systems? *Waste Management*, **26(5)**, pp. 516–525, 2006. [doi:http://dx.doi.org/10.1016/j.wasman.2005.07.021](http://dx.doi.org/10.1016/j.wasman.2005.07.021)
- [7] Haubrichs, R. & Widmann, R., Evaluation of aerated biofilter systems for microbial methane oxidation of poor landfill gas. *Waste Management*, **26(4)**, pp. 408–416, 2006. [doi:http://dx.doi.org/10.1016/j.wasman.2005.11.008](http://dx.doi.org/10.1016/j.wasman.2005.11.008)
- [8] Scheutz, C., Kjeldsen, P., Bogner, J.E., De Visscher, A., Gebert, J., Hilger, H.A., Huber-Humer, M. & Spokas, K., Microbial methane oxidation processes and technologies for mitigation of landfill gas emissions. *Waste Management and Research*, **27(5)**, pp. 409–455, 2009. [doi:http://dx.doi.org/10.1177/0734242X09339325](http://dx.doi.org/10.1177/0734242X09339325)
- [9] Hanson, R. & Hanson, T., Methanotrophic bacteria. *Microbiological Reviews*, **60(2)**, pp. 439–471, 1996.
- [10] Humer, M. & Lechner, P., Alternative approach to the elimination of greenhouse gases from old landfills. *Waste Management and Research*, **17(6)**, pp. 443–452, 1999.
- [11] Zeiss, C.A., Accelerated methane oxidation cover system to reduce greenhouse gas emissions from MSW landfills in cold, semi-arid regions. *Water, Air, and Soil Pollution*, **176(1–4)**, pp. 285–306, 2006.
- [12] Scheutz, C. & Kjeldsen, P., Environmental factors influencing attenuation of methane and hydrochlorofluorocarbons in landfill cover soils. *Journal of Environmental Quality*, **33(1)**, pp. 72–79, 2004. [doi:http://dx.doi.org/10.2134/jeq2004.0072](http://dx.doi.org/10.2134/jeq2004.0072)
- [13] Visvanathan, C., Pokhrel, D., Cheimchaisri, W., Hettiaratchi, J.P.A. & Wu, J.S., Methanotrophic activities in tropical landfill cover soils: effects of temperature, moisture content and methane concentration. *Waste Management and Research*, **17(4)**, pp. 313–323, 1999.
- [14] Park, S., Lee, C., Ryu, C. & Sung, K., Biofiltration for reducing methane emissions from modern sanitary landfills at the low methane generation stage. *Water, Air, and Soil Pollution*, **196(1–4)**, pp. 19–27, 2009.
- [15] Einola, J.-M., Sormunen, K.M. & Rintala, J.A., Methane oxidation in a boreal climate in an experimental landfill cover composed from mechanically-biologically treated waste. *Science of the Total Environment*, **407(1)**, pp. 67–83, 2008. [doi:http://dx.doi.org/10.1016/j.scitotenv.2008.08.016](http://dx.doi.org/10.1016/j.scitotenv.2008.08.016)
- [16] Mudliar, S., Giri, B., Padoley, K., Satpute, D., Dixit, R., Bhatt, P., Pandey, R., Juwarkar, A. & Vaidya, A., Bioreactors for treatment of VOCs and odours – a review. *Journal of Environmental Management*, **91(5)**, pp. 1039–1054, 2010.
- [17] Gebert, J., Groengroeft, A. & Miehlich, G., Kinetics of microbial landfill methane oxidation in biofilters. *Waste Management*, **23(7)**, pp. 609–619, 2003. [doi:http://dx.doi.org/10.1016/S0956-053X\(03\)00105-3](http://dx.doi.org/10.1016/S0956-053X(03)00105-3)
- [18] Kallistova, A.Y., Kevbrina, M.V., Nekrasova, V.K., Shnyrev, N.A., Einola, J.M., Kulomaa, M.S., Rintala, J.A. & Nozhevnikova, A.N., Enumeration of methanotrophic bacteria in the cover soil of an aged municipal landfill. *Microbial Ecology*, **54(4)**, pp. 637–645, 2007. [doi:http://dx.doi.org/10.1007/s00248-007-9219-0](http://dx.doi.org/10.1007/s00248-007-9219-0)

- [19] Maestre, J.P., Gamisans, X., Gabriel, D. & Lafuente, J., Fungal biofilters for toluene biofiltration: evaluation of the performance with four packing materials under different operating conditions. *Chemosphere*, **67**(4), pp. 684–692, 2007. [doi:http://dx.doi.org/10.1016/j.chemosphere.2006.11.004](http://dx.doi.org/10.1016/j.chemosphere.2006.11.004)
- [20] Leson, G. & Winer, A., Biofiltration – an innovative Air-Pollution Control Technology for VOC emissions. *Journal of the Air & Waste Management Association*, **41**(8), pp. 1045–1054, 1991.
- [21] Nikiema, J., Bibeau, L., Lavoie, J., Brzezinski, R., Vigneux, J. & Heitz, M., Biofiltration of methane: an experimental study. *Chemical Engineering Journal*, **113**(2–3), pp. 111–117, 2005.
- [22] Phillips, R.L., Whalen, S.C. & Schlesinger, W.H., Influence of atmospheric CO₂ enrichment on methane consumption in a temperate forest soil. *Global Change Biology*, **7**(5), pp. 557–563, 2001. [doi:http://dx.doi.org/10.1046/j.1354-1013.2001.00432.x](http://dx.doi.org/10.1046/j.1354-1013.2001.00432.x)
- [23] Spokas, K.A. & Bogner, J.E., Limits and dynamics of methane oxidation in landfill cover soils. *Waste Management*, 2010.
- [24] Alagappan, G. & Cowan, R.M., Effect of temperature and dissolved oxygen on the growth kinetics of *Pseudomonas putida* F1 growing on benzene and toluene. *Chemosphere*, **54**(8), pp. 1255–1265, 2004.
- [25] Esener, A.A., Roels, J.A. & Kossen, N.W.F., The influence of temperature on the maximum specific growth rate of *Klebsiella pneumoniae*. *Biotechnology and Bioengineering*, **23**, pp. 1401–1405, 1981. [doi:http://dx.doi.org/10.1002/bit.260230620](http://dx.doi.org/10.1002/bit.260230620)
- [26] Streese, J., Schlegelmilch, M., Heining, K. & Stegmann, R., A macrokinetic model for dimensioning of biofilters for VOC and odour treatment. *Waste Management*, **25**(9), pp. 965–974, 2005. [doi:http://dx.doi.org/10.1016/j.wasman.2005.07.009](http://dx.doi.org/10.1016/j.wasman.2005.07.009)
- [27] Sologar, V.S., Lu, Z. & Allen, D.G., Biofiltration of concentrated mixtures of hydrogen sulfide and methanol. *Environmental Progress*, **22**(2), pp. 129–136, 2003. [doi:http://dx.doi.org/10.1002/ep.670220215](http://dx.doi.org/10.1002/ep.670220215)
- [28] Avalos Ramirez, A., Benard, S., Giroir-Fendler, A., Jones, J.P. & Heitz, M., Kinetics of microbial growth and biodegradation of methanol and toluene in biofilters and an analysis of the energetic indicators. *Journal of Biotechnology*, **138**(3–4), pp. 88–95, 2008.
- [29] Arrhenius, S., Über die Reaktionsgeschwindigkeit bei der Inversion von Rohrzucker durch Säuren. *Zeitschrift für Physikalische Chemie*, **4**, pp. 226–248, 1889.
- [30] Esener, A.A., *An Engineering Study of Bacterial Kinetics and Energetics*, Delft University Press, 1981.
- [31] Nesbit, S. & Breitenbeck, G., A laboratory study of factors influencing methane uptake by soils. *Agriculture Ecosystems & Environment*, **41**(1), pp. 39–54, 1992. [doi:http://dx.doi.org/10.1016/0167-8809\(92\)90178-E](http://dx.doi.org/10.1016/0167-8809(92)90178-E)
- [32] Whalen, S.C., Reeburgh, W.S. & Sandbeck, K.A., Rapid methane oxidation in a landfill cover soil. *Applied and Environmental Microbiology*, **56**, pp. 3405–3411, 1990.
- [33] Nozhevnikova, A.N., Nekrasova, V.K., Kevbrina, M.V. & Kotsyurbenko, O.R., Production and oxidation of methane at low temperature by the microbial population of municipal sludge checks situated in north-east Europe. *Water Science and Technology*, **44**(4), pp. 89–95, 2001.
- [34] de Ory, I., Romero, L.E. & Cantero, D., Modelling the kinetics of growth of *Acetobacter aceti* in discontinuous culture: influence of the temperature of operation. *Applied Microbiology and Biotechnology*, **49**(2), pp. 189–193, 1998. [doi:http://dx.doi.org/10.1007/s002530051157](http://dx.doi.org/10.1007/s002530051157)