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**Methane oxidation in a biofilter (part 2):
a lab-scale experiment for model calibration**

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Abstract

Methane is a strong greenhouse gas, 21 times more than carbon dioxide. Landfills are one of the most important anthropogenic source of methane through biogas production. For small or old landfills it is not possible to install energy recovery systems and in several cases it is impossible to use flares as well. In these situations biofiltration represents a valid alternative for biogas treatment. In biofilters methane is oxidized by methanotrophic bacteria. In this study an experimental study on a biological methane oxidation column presented with the aim to calibrate a mathematical model developed in an earlier study. The column was designed to reproduce at lab-scale a real biofilter trying to consider the more probable landfill boundary conditions. Although the methane oxidation efficiency in the column was lower than the expected (around 35%), an appropriate model implementation showed an acceptable agreement between the outcomes data of the model simulation and the experimental data (with a Theil's Inequality Coefficient value of 0.08). A calibrated model allows a better management of the biofilter performance in terms of methane oxidation.

Keywords: Methane oxidation, methane emission, landfill gas, *methanotrophs* *microorganisms*, biofilters.

Introduction

Biogas treatment

The methane is an important greenhouse gas: 21 times more effective at trapping heat than the carbon dioxide ^[1]. In the last years the methane concentration in the atmosphere increased from a value of 715 parts per billion (ppb) related to pre-industrial age to 1774 ppb registered in 2005 ^[2]. One of the most important anthropogenic sources of the methane is biogas that is produced by the anaerobic decomposition of organic waste. The main components of this biogas are indeed carbon dioxide (CO₂) and methane (CH₄). In the US about 6.329 Tg of CH₄ were produced from landfills in 2007 ^[3]. In Europe, in 2006, the methane from landfills was the second largest source of anthropogenic CH₄ (22%) with an estimated CH₄ emission of 3.373 Tg from waste disposal ^[4]. Worldwide landfills produce large amount of biogas. Some studies tried to calculate the values of methane released in atmosphere every year by landfills worldwide and this value is between 35 and 69 Tg year⁻¹ of CH₄ ^[5].

In relation with the environmental problematics related to the global warming, in the last few years many countries have recognized the treatment of biogas as an important step to lower the emission of greenhouse gasses. In all the developed countries new landfills must provide for a biogas extraction system linked with an energy recovery plant.

However there are different situations, such as small waste disposal sites or old landfills in which is not possible or not cost-effective making a new LFG (landfill gas) extraction system or a new energy recovery plant. For these situations there are different way to treat the biogas in order to lower the methane concentration content in the flow.

Flares represent a system to reduce the methane present in the biogas. The methane is oxidized to carbon dioxide and water through combustion. Technical specifications for flares available on the market indicate a minimum flow of 50 Nm³/h and a methane concentration of 30% v/v ^[6]. If these parameters are not respected the incomplete combustion would lead the production

of other gaseous pollutants (hydrocarbons, nitrogen oxides, carbon monoxide etc.). In many cases the flow and the methane concentration in the gas are low for any flares.

The biofiltration

The biofiltration is a biologic treatment for the biogas and represent a reliable system for those situations characterized by low flow quantity (less than 50 Nm³/h) and low methane concentration (less than 30%). The process consist in a biologic oxidation occurred by the methanotrophic bacteria. These bacteria are able to use the methane content in the gas flow as carbon source. The biochemical reaction ($\Delta^\circ G = -780 \text{ kJ mol}^{-1} \text{ CH}_4$, $\Delta^\circ H = -890 \text{ kJ mol}^{-1}$) is the following [7-8]:

The kinetics of the process are described by *Michaelis–Menten* [8-9-10]:

$$r = \frac{V_{\max} \cdot C_{\text{CH}_4}}{K_m^{\text{CH}_4} + C_{\text{CH}_4}} \cdot \frac{C_{\text{O}_2}}{K_m^{\text{O}_2} + C_{\text{O}_2}}$$

Where:

- $r = \text{CH}_4 \text{ oxidation rate } [\text{mol}/(\text{m}^3 \cdot \text{h})];$
- $V_{\max} = \text{maximum CH}_4 \text{ oxidation rate } [\text{mol}/(\text{m}^3 \cdot \text{h})];$
- $K_m = \text{Michaelis–Menten or half-saturation constant } [\text{mol}/\text{m}^3];$
- $C = \text{concentration } [\text{mol}/\text{m}^3];$

The biofilter is an aerobic reactor in which the biochemical reaction described before is occurring. Generally biofilters are containers filled with material that allows the methanotrophic bacteria to develop. The filling material is the heart of the system and different studies were focused to find the best composition to oxidize the methane [11-12-13-14]. Wood chips are considered a good material to support and sustain the development of a methanotrophic biofilm. From a technical point of view the LFG is conveyed and conducted by

a piping system to the biofilter ^[15]. To avoid preferential pathways in the bottom of the container there is an empty space in which the LFG can be distributed uniformly over the entire oxidation surface.

As for all the biological processes many environmental factors influence the methane oxidation efficiency. The temperature is an important parameter that control this process ^[16]. The optimal range for these bacteria is around 25 – 35 °C ^[17], while 31°C is mentioned as the optimal temperature by King & Adamsen ^[18], methane oxidation can occur down to 1–2 °C ^[19]. The moisture of the filling material is important for the nutrient transport and it has to be between 10-20% w/w ^[20]. If the water content is higher the methane oxidation efficiency goes down (molecular diffusion in water is about 100 times slower than in the air ^[21]). If this value is under the threshold value of 5% the bacteria activity is drastically reduced as the water stress in the system influences the nutrient supply ^[22-23]. The pH value for the methane oxidation must be between 5.5 and 8.5 ^[10]. The exopolymeric substances (EPS) can also influence the methane oxidation efficiency. EPS accumulation in the biofilm reduces the CH₄ oxidizing capacity ^[24-25].

Aim of the study

The biogas treatment for small or landfills in advanced stages of post management is a challenging task. The biofiltration is a good solution because it is not affected by the low LFG flow or by the low methane concentration in the biogas. The development of a mathematical model to manage and control the biofiltration process is very important in order to optimize the performance of a biofilter. The aim of this study concerns the calibration and use of the mathematical model developed and tested by Amodeo et al. ^[26] through a lab-experiment, considering the geometric characteristics of the lab-scale column.

Materials and methods

For the process of model calibration a lab-scale column was constructed (Fig. 1).

The column was made in glass with a diameter of 7 cm (resulting in a horizontal surface of 0.0038 m^2). The total height was 95 cm divided in: (from the bottom) 5 cm of free space in order to have a good distribution of the gas, 10 cm of gravel to avoid preferential pathways for the gas phase, 70 cm of filling material; 10 cm of free space to allow a good mixing of the exhaust gas and also to insert all the instruments such as temperature sensor, water input and gas output pipe. Between the gravel and the free space a plexiglass filter of 5 mm was placed to prevent the gravel from falling down. To close the column in the top a cork stopper was used with three holes (one for the temperature sensor, one for the water input pipe and one for the gas output pipe).

Filling material

The aim of the work was to reproduce the conditions of a full scale biofilter. As suggested in literature ^[27] the material selected for this work was a 50% - 50% mixture of compost and landfill soil. The presence of compost ensures the organic substance, necessary for the biologic processes (in this case for the methanotrophic bacteria development). Due to exposure to any biogas losses, the landfill soil is very important as well, because it could already contain several methanotrophic bacterial cultures. The landfill soil was taken from Vanheede landfill (municipality of Roeselare, Belgium, www.vanheedelandfillsolutions.be). The mixing soil had a specific weight of 645.6 kg/m^3 , a relative moisture content of 17.82% and a total solids content of 82.18%, determined by drying 3 soil samples of 10 g each for 48 h at 105°C .

Flow and gas composition

The gas composition and the flow are very important parameters ^[28]. The goal of this work is to reproduce as much as possible the functioning of a full scale biofilter. The landfill operative

conditions, for which the biofiltration represent an interesting solution, are with low methane content in the biogas (about 10%). For this reason a gas flow with 3% of methane was used. This value is lower than 10% for safety reasons (the experiment was conducted indoors). Further, the gas contained 20% of oxygen (according to Wang et al. [29]) and 77% of nitrogen. This gas composition was purchased as such at Air Liquide. The applied flow rate it was 0.1 liter per minute (lpm). This value came out considering, as usual in this work, the operative conditions in small and/or old landfills. A full scale biofilter is designed for flow of around 50 Nm³/h [30]. If it is considered that the ratio between the volume of the biomass in the column (0.0027 m³) and the biomass volume in the full scale biofilter (22 m³) is 0.0012, a flow rate of 0.1 lpm was obtained ($50 \text{ Nm}^3/\text{h} \cdot 0.0012 = 0.006 \text{ Nm}^3/\text{h} = 0.1 \text{ lpm}$). Now if the total flow (0.1 lpm), the methane percentage in the flow (3%) and the effective oxidation surface of the column (0.0038 m²) are known it is possible to calculate the value of the methane loading rate:

which is $50 \frac{\text{mol}_{\text{CH}_4}}{\text{m}_{\text{soil}}^2 \cdot d}$.

Experimental procedures

After the column was filled with the soil, the gas was introduced from the bottle to the column. The experiment was conducted for two months. During the first part (one month) the gas, before entering in the column, was directed in a gas-washing bottle filled with water in order to saturate the gas. In this way it was tried to avoid that the gas took away moisture from the soil. The experiment in this first part was conducted with an on/off system: the gas flow was on for 8 hours per day and 5 days per week. During the second part of the experiment (another month) the gas was conducted directly to the column (without saturation) and the system was operated continuously (24 hours per day 7 days per week). Every day several parameters were monitored. Soil temperature was measured (two times per day) with a *Traceable*®

Thermometer with a sensor of 30 cm and a range of -50 - 150 °C. To measure the flow a *Cole Parmer*® flowmeter was installed with (measurement range 0 – 0.5 lpm).

Gas samples and gas analysis

A valve system has been prepared for collecting the exhaust gas samples from the top of the column. During the experiment this valve system provided a possibility to directly sample the gas from the column, as during sample collection all the gas flow is directed in a tube already connected with a 12 mL syringe. The gas collected in the syringes was transferred in small bottles sealed with rubber stoppers. The bottle was filled with an acid solution composed of sulfuric acid, distilled water and methyl orange. The pH of the solution was around 2 (generally the ratio is 200 µl of sulfuric acid for liter of distilled water). The aim of the solution is to retain the gas and in the same time to avoid gas transfer to the liquid phase. It is only possible that a portion of carbon dioxide is transferred from the biogas to the solution, although the low pH minimizes this effect. At the same time, for each sample of exhaust gas, an input gas sample was taken in the same way.

The gas samples were analyzed using the gas chromatograph *Agilent 6890 Serie Plus*. A Hamilton gas syringe (250 µL) was used. The injector temperature was 65°C, the oven temperature was 55°C and the temperature of the Flame Ionization detector (FID) was 200°C. The chromatograph was calibrated with a known gas composition (80% of methane, 10% of carbon dioxide, 5% of nitrogen and 5% of hydrogen).

Mathematical model description

The mathematical model considered was developed by Amodeo et al. ^[26]. The model calculates the methane, carbon dioxide and oxygen concentration as function of the filter height as function of for example the biogas flow. As such the following equations were used:

$$(1) \quad \frac{dy_{CH_4}}{d\omega} = -St_{CH_4} \cdot (y_{CH_4} - H_{CH_4} \cdot y_{CH_4}^S)$$

$$y_{CH_4}^S = \frac{-\left(\frac{X_{CH_4}}{H_{CH_4}} - \frac{y_{CH_4}}{H_{CH_4}} + K_m^{CH_4}\right) + \sqrt{\left(\frac{X_{CH_4}}{H_{CH_4}} - \frac{y_{CH_4}}{H_{CH_4}} + K_m^{CH_4}\right)^2 + 4 \frac{K_m^{CH_4} y_{CH_4}}{H_{CH_4}}}}{2}$$

$$(2) \quad \frac{dy_{O_2}}{d\omega} = -St_{O_2} \cdot (y_{O_2} - H_{O_2} \cdot y_{O_2}^S) \quad y_{O_2}^S = \frac{y_{O_2}}{H_{O_2}} - 2 \cdot \frac{X_{O_2}}{H_{O_2}} \cdot \frac{y_{CH_4}^S}{K_m^{CH_4} + y_{CH_4}^S}$$

$$(3) \quad \frac{dy_{CO_2}}{d\omega} = St_{CO_2} \cdot (y_{CO_2} - H_{CO_2} \cdot y_{CO_2}^S) \quad y_{CO_2}^S = \frac{y_{CO_2}}{H_{CO_2}} - \frac{X_{CO_2}}{H_{CO_2}} \cdot \frac{y_{CH_4}^S}{K_m^{CH_4} + y_{CH_4}^S}$$

Where:

- $y_{CH_4}, y_{O_2}, y_{CO_2}$ = mole fraction in the gas phase [dimensionless];
- $\omega = z / h$, with h the height of the filling material and z the axial coordinate
- $St_{CH_4}, St_{O_2}, St_{CO_2}$ = Stanton number [dimensionless];
- $H_{CH_4}, H_{O_2}, H_{CO_2}$ = Henry's law constant [dimensionless];
- $y_{CH_4}^S, y_{O_2}^S, y_{CO_2}^S$ = mole fraction on the biomass boundary [dimensionless];
- $X_{CH_4}, X_{O_2}, X_{CO_2}$ = ratio between Damköhler and Stanton number [dimensionless];
- $K_m^{CH_4}, K_m^{O_2}, K_m^{CO_2}$ = half-saturation constant [dimensionless];

For the nominal operating conditions (Tab.1) of an aerobic full scale biofilter the results of the model are shown in the Figure 2.

Results and Discussion

The results of the lab-scale experimental column show that the methane oxidation occurred by the methanotrophic bacteria. These results were compared with those obtained from the mathematical model in order to perform a calibration.

Model Implementation

For simulations with the mathematical model the following influent conditions and model parameters were used as described in the material and methods section and summarised in Table 2.

The value of the maximum oxidation rate V_{max} is very important for the methane oxidation efficiency [26-31-32]. After several considerations it was decided to initially use 4 mol/(m³*h) as a starting value, considering that the maximum value of V_{max} is 8 mol/(m³*h) and the minimum is 0 as reported by De Visscher & Van Cleemput [33].

Methane oxidation profiles

First it is important to notice that the methane loading rate of the column (described in the paragraph 2.2) is a high value in relation with the values used in literature (Tab. 3) in order to keep in lab-scale the operative conditions in small and/or old landfills (Abichou et al. [34] show values of V_{max} measured on different soils obtained from the available literature).

Figure 3 shows an overview of the results considering 8 samples. The lines represent the results predicted by the mathematical model while the points represent the results obtained experimentally in the column. In this way, considering the methane oxidation efficiency (defined in Hettiarachchi et al. [37]), it's possible to note the comparison between the model simulations and the column experiment. By using a V_{max} value of 4 mol/(m³*h), the methane oxidation was overestimated. As such, a value of 2.5 mol/(m³*h) was considered.

The continues line was determined using a V_{max} value of 4 mol/(m³*h), the dotted line was determined with a V_{max} value of 2.5 mol/(m³*h). This parameter is the most influential parameter in the model, as was indicated by a sensitivity analysis [26].

The graph shows that the correlation between the model and the experiment is better with a V_{max} value of 2.5 mol/(m³*h). To evaluate numerically the goodness-of-fit between experimental and simulated values the *Theil's Inequality Coefficient* (TIC) was considered [38]:

$$TIC = \frac{\sqrt{\sum_i (y_i - y_{m,i})^2}}{\sqrt{\sum_i y_i^2} + \sqrt{\sum_i y_{m,i}^2}}$$

Where y represents the values predicted by the mathematical model and y_m the value measured from the lab-column. If the value of the TIC is lower than 0.3 the correlation between the model and experiment data is good. In this case the calculation gave a TIC value of 0.14 for $V_{max} = 4 \text{ mol}/(\text{m}^3 \cdot \text{h})$ and 0.08 for $V_{max} = 2.5 \text{ mol}/(\text{m}^3 \cdot \text{h})$. Both the situations are acceptable but the model simulation with $V_{max} = 2.5 \text{ mol}/(\text{m}^3 \cdot \text{h})$ fits better.

From a theoretical point of view the use of a lower value of V_{max} means that the biological activity is not so good as the prediction. The reasons could be the initial intermittent flow, the possibility that the moisture is not included in the correct range (10-20%), the probably not fully developed methanotrophic biomass and the high methane loading rate.

Conclusions

The aim of the work was the use of data from a lab-scale column for the calibration of a mathematical model. The results show that the methane oxidation in the column occurs with a methane oxidation efficiency comparable with literature values [20-31-32-39].

The methane oxidation, made by methanotrophic bacteria, is a biologic phenomenon: many environmental factors can occur, so it is very hard to find a perfect correspondence between the model prediction and the measured values. There is a difference between both values but the numeric value of the TIC suggests a good calibration of the model.

The next step will be the testing of the model with data from a full scale biofilter.

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FIGURE CAPTIONS

Figure 1. Schematic diagram of the experimental column

Figure 2. Variation of the mole fraction as a function of the biofilter height

Figure 3. Methane oxidation efficiency measured in the column and predicted with the model

Table 1. Nominal operating conditions of the aerobic biofilter

Parameters	Values	UM
Length	6.5	m
Width	2.5	m
Height	1.4	m
Surface	16.25	m ²
Volume	22.75	m ³
Q	50	Nm ³ /h
T	30	°C
C _{TOT.}	43.8	mol/m ³
CH ₄	20	%
CO ₂	20	%
O ₂	21	%

Table 2. Parameters and influent conditions included in the model

Parameters	Value	Unit
Soil height	70	cm
Column diameter	7	cm
Oxidation surface	38.46	cm ²
Column volume	2692.55	cm ³
Max. oxidation rate $V_{\max}$	2,5-4	mol/(m ³ *h)
Influent	Value	Unit
Flow – Q	0.1	lpm
Soil temperature – T	variable	°C
CH ₄ molar fraction	3	%
CO ₂ molar fraction	0	%
O ₂ molar fraction	20	%
N ₂ molar fraction	77	%

Table 3. Methane mass flow rate considered

Reference	Value	UM
De Visscher et al. ^[31]	12.5	mol / m ² *d
Stein et al. ^[32]	19.7	mol / m ² *d
Park et al. ^[20]	30	mol / m ² *d
De Visscher et al. ^[35]	13.4	mol / m ² *d
Mahieu et al. ^[36]	19.3	mol / m ² *d
<i>Amodeo et al.</i>	50	mol / m ² *d