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**Methane oxidation in a biofilter (part 1):
development of a mathematical model for designing and optimization**

CORRADO AMODEO^{1,2}, SALVATORE MASI¹, STIJN W.H. VAN HULLE², PIERFRANCESCO
ZIRPOLI¹, IGNAZIO M. MANCINI¹ and DONATELLA CANIANI¹*

¹ *School of Engineering, University of Basilicata – Viale dell’Ateneo Lucano - 85100 Potenza (Italy)*

² *Department of Industrial Biological Sciences, Ghent University Campus Kortrijk, Graaf Karel de
Goedelaan, 5 - 8500 Kortrijk (Belgium)*

*Address correspondence to Corrado Amodeo, Department of Industrial Biological Sciences, Ghent University Campus Kortrijk, Graaf Karel de Goedelaan, 5 - 8500 Kortrijk (Belgium); Phone: +32 493673719; Fax: +32 56241224;

E-mail: corrado.amodeo@unibas.it

Abstract

Landfill gas is produced by anaerobic degradation of organic waste. Landfills are one of the principal anthropogenic sources of atmospheric methane, a strong greenhouse gas ^[1]. Landfills, in particular, have been found to be a principal source of CH₄ production. The CH₄ emission from the waste sector is about 18% of the global anthropogenic CH₄ emission ^[2]. For small landfills or landfills in advanced stages of post management (with methane percentage below 30% v/v) biofilters are an interesting sustainable alternative. The aim of this work is the evaluation of the efficiency of such a biofilter, through the application of a mathematical model which describes the biological oxidation process. This mathematical model is able to predict the efficiency of the system under varying operating conditions. Literature data have been used in order to build the model. The factors that mostly affect the process and which actually regulate the entire process have been highlighted in this work. Specifically, it was found that temperature, flow and methane concentration are the most important parameters that influence the system. The results obtained from the mathematical model showed also that the biofilter system is simple to implement and manage and allows the achievement of high efficiency of methane oxidation. In the optimal conditions for temperature (between 20-30 °C), residence time (between 0.7-0.8 h) and methane molar fraction (between 20-25%) the efficiency of methane oxidation could be around 50%.

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

Introduction

Landfill gas (LFG)

A series of control and abatement systems must be provided to minimize the environmental impact that may derive from landfills (during the management or during the post-management). In the landfills the decomposition processes of organic substances under anaerobic conditions lead to the production of biogas which is usually composed mainly of methane (CH_4) and carbon dioxide (CO_2) ^[3]. Both are classified as greenhouse gases. In the atmosphere the concentration of CH_4 has increased from a pre-industrial value of about 715 to 1732 parts per billion (ppb) in the early 1990s, and was 1774 ppb in 2005. The atmospheric concentration of CH_4 in 2005 far exceeded the natural range of the last 650 000 years (320 to 790 ppb) as determined from ice cores ^[4]. In the last years the values of methane concentration are due to continuing anthropogenic CH_4 emissions. Several studies, such as Forester et al. ^[1] have demonstrated that the contribution of methane to climate change is 18% of the total radiative forcing by all long-lived greenhouse gases. In the United States the methane originated from landfills is the second largest anthropogenic CH_4 emission source: the emission of CH_4 from US landfills in 2007 was about 6.329 Tg ^[5]. Also in Europe, in 2006, the methane from landfills was the second largest source of anthropogenic CH_4 (22%) with an estimated CH_4 emission of 3.373 Tg from waste disposal ^[6]. If the contribution of all the landfills in the world is considered, the amount of methane released is between 35 and 69 Tg year⁻¹ of CH_4 ^[2]. Obviously all these data do not originate from measurements on site but are projections based on estimated rates of CH_4 production in relation with the national statistics about (landfilled) waste ^[3].

LFG treatment

The contribution of CH_4 to global GHG emissions is important and for this reason worldwide most of legislative systems provide several treatments for the municipal waste before or in alternative to the landfill disposal ^[7]. As a consequence of the large contribution of LFG to the global greenhouse

gas production, most of the industrialized countries have imposed legislation for the extraction and treatment of landfill gas, especially in recently constructed installations.

In the large and young landfills the energy recovery systems represent the best solution to treat the landfill gas but it need large quantity of LFG flow rate and a high enough methane content. Flares are another solution to treat the biogas, oxidizing CH_4 through combustion. Technical specifications for the operation of gas flares available on the market indicate a minimum flow of $50 \text{ Nm}^3/\text{h}$ and a methane concentration of 30% v/v (as is shown in a normal catalogue of flares ^[8]). When the flow is less than $50 \text{ Nm}^3/\text{h}$ or the methane concentration of the biogas is less than 30% v/v the biofiltration represent a good way to treat the biogas.

The biofilter is an aerobic reactor that can be used for treatment of landfill gas by methane oxidation, so called microbial aerobic methane oxidation (MAMO). The biofilter contains microorganisms able to convert the methane to carbon dioxide ^[3]. In the biofilter the filling material supports and sustains the development of a methanotrophic biofilm. As usual the biofiltration costs (both investment and management) decrease relatively with increasing size of the biofilter. The average investment costs of a biofilter of 10, 20, and 40 m^3 are € 1300, € 1100, and € 800 per m^3 of reactor, respectively. The operational costs, including both fixed and variable costs, are € 190, € 160, € 124 per m^3 of reactor per year, respectively ^[9].

There are two different biofilter systems: upflow or downflow. The upflow system, if it work as an open bed, allows the presence of atmospheric oxygen. The extraction of LFG may be active or passive. For the active system, the LFG extracted from the landfill is supplied to the biofilter and so it is possible to control the influent flow. Air and LFG can be combined before entering in the system ^[10-11], or two different feed lines can be used ^[12]. Air may also enter in the biofilter by diffusive transport across the column surface ^[13]. The systems with active feeding works usually with constant temperature and water content and so a power supply is needed. It is more complicate to manage this kind of systems so often these systems are housed to facilitate the control over the operational parameters. On the other side, in the passive systems, the LFG comes in when there is a

pressure gradient between the landfill and the atmosphere. In these conditions the gas passes through collection piping up to arrive in the biofilter ^[14-15-16]. When the pressure gradients are not high the diffusion process may play an important role, while the O₂ is supplied to the system or by advection when the pressure rises or by diffusion from the atmosphere across the biofilter surface ^[16]. So the weak point of passive systems is that the operating conditions are not manageable, but on the other side all the costs (capital and operating) are considerably lower in relation with active systems^[3].

Methane oxidation kinetics

The process of microbial aerobic methane oxidation (MAMO), according to Chanton et al. ^[17] and Ng et al. ^[18], is regulated by the following biochemical reaction ($\Delta^\circ G = -780 \text{ kJ mol}^{-1} \text{ CH}_4$, $\Delta^\circ H = -890 \text{ kJ mol}^{-1}$):

Where x is the stoichiometric coefficient. The kinetics that occurs is described by Michaelis–Menten kinetics for methane and oxygen ^[3-18-19]:

$$r = \frac{V_{\max} \cdot y_{\text{CH}_4}}{K_m^{\text{CH}_4} + y_{\text{CH}_4}} \cdot \frac{y_{\text{O}_2}}{K_m^{\text{O}_2} + y_{\text{O}_2}} \quad (1)$$

Where:

- $r = \text{CH}_4 \text{ oxidation rate } [\text{mol}/(\text{m}^3 \cdot \text{h})]$;
- $V_{\max} = \text{maximum CH}_4 \text{ oxidation rate } [\text{mol}/(\text{m}^3 \cdot \text{h})]$;
- $K_m = \text{Michaelis–Menten (or half-saturation) constant } [\text{dimensionless}]$;
- $y_{\text{CH}_4} = \text{molar fraction of CH}_4 \text{ in the biofilm} [\text{dimensionless}]$;
- $y_{\text{O}_2} = \text{molar fraction of O}_2 \text{ in the biofilm } [\text{dimensionless}]$.

The methanotrophic bacteria are aerobic bacteria, so require the presence of oxygen in order to convert methane into carbon dioxide. The oxygen concentration is an important parameter: the

efficiency of methane oxidation is near the maximum when oxygen concentration reached 5% v/v, and then keep a stable value when it reached 21% v/v ^[20]. If the concentration is under this threshold there will be a rapid decrease of the conversion rates of methane.

As all the biological reaction there are several environmental factors that control the methane oxidation (e.g soil texture, temperature, soil moisture content, methane and oxygen supply, nutrients, etc.). Thus, all the environmental conditions are very important for the methane oxidation rate and should therefore be included in the kinetic model. A main role is played by the temperature that has a profound effect on all biological processes, including methane oxidation activity. Most of methanotrophs bacteria are mesophiles so their optimum temperature is around 25 – 35 °C ^[3]. However, methane oxidation can occur down to 1–2 °C ^[21].

Another important factor for micro-organisms is certainly the moisture, as transport medium for nutrient supply and also for removal of residual metabolic compounds ^[22]. So the efficiency of the methane oxidation depends of the moisture contents of the system: low and high water contents can limit the process. If the moisture is high the gaseous transport processes in the soil may slow down (molecular diffusion in water is about 10^4 times slower than in the air ^[23]). Indeed, if the soil's degree of saturation (that represent the ratio between the volume of water and the volume of voids) reaches high values (e.g. 85%), the air filled voids are no longer interconnected. In these conditions the gases must diffuse in the liquid phase, and this means that the methane and oxygen availability is drastically reduced, limiting the oxidation process. On the other side if the moisture content decreases the oxidation efficiency may also reduce significantly due to water stress in the system. When the soil conditions are optimum in term of moisture content the oxidation process can occur with good results. The specific value obviously is function of the specific characteristics of the biofilter; generally in the range between 10-20% w/w. The threshold value is 5%: if the moisture content drops below this value then the oxidation activity is drastically reduced.

Another important parameter for the success of the process that plays an important role is the pH, as in the most of reactions. The optimal pH for the methane oxidation and so for the methanotrophic bacteria development is between 5.5 and 8.5 ^[3].

MAMO modelling

Bogner et al. ^[24] developed a dynamic model for landfill gas transport and methane oxidation in landfill cover soils in terms of molecule collisions with the soil matrix. Hilger et al. ^[25] developed a model based on the well-established Stefan-Maxwell equations. Stein et al. ^[26] developed a model for simulating 1D transport and biological reaction of methane in porous media. De Visscher et al. ^[27] developed a simulation model that describes gas transport and methane oxidation in a laboratory setup. Hettiarachchi et al. ^[28] and Hettiarachchi et al. ^[29] did a comprehensive one-dimensional mathematical model that incorporates heat transfer and moisture transport in methane biofilters. Ng et al. ^[18] developed a model that incorporates water, gas and heat coupled reactive transport in unsaturated soil and Abichou et al. ^[30] used a numerical model that combines flow of water and heat with gas transport. Such models provide a practical tool for prediction of net methane conversion and they also allow a sensitivity analysis of the most important parameters. The main difference between the model presented in this study and existing models is that most of these models refer to the process of methane oxidation that occurs in landfill covers and not in biofilters. In the latter case all the gases enter from the bottom of the system (already premixed). Other differences are the choice of diffusion coefficients to explain the transport phenomena and the use of two dimensionless numbers to solve the model equations. Therefore, in this study, a novel model was developed.

Aim of the study

The aim of this work is the evaluation of the efficiency of a biofilter, through the application of the above mentioned novel mathematical model which incorporates the effects on the biological

oxidation process. This mathematical model will be used to give some practical information to design and manage a biofilter for landfills.

Materials and methods

Reaction and reactor model

The simplifying assumption for the implementation of the mathematical model (in Matlab®) is that the biofilter is an ideal plug flow reactor (PFR) with complete mixing in the transverse direction and that the gas mixture consists of only CH₄, O₂, CO₂ and N₂. All the gases, including oxygen, enter from the bottom of the biofilter (*Fig. 1*). The nitrogen was not considered in the model (as it is inert in MAMO).

For the three components (CH₄, O₂, CO₂) a steady state mass balance was set-up.

$$\frac{dy_{CH_4}}{dz} = -\frac{k_g^{CH_4} a}{Q} (y_{CH_4} - H_{CH_4} \cdot y_{CH_4}^s) \cdot S \quad (2)$$

$$\frac{dy_{O_2}}{dz} = -\frac{k_g^{O_2} a}{Q} (y_{O_2} - H_{O_2} \cdot y_{O_2}^s) \cdot S \quad (3)$$

$$\frac{dy_{CO_2}}{dz} = \frac{k_g^{CO_2} a}{Q} (y_{CO_2} - H_{CO_2} \cdot y_{CO_2}^s) \cdot S \quad (4)$$

With:

- Q = landfill flow rate entering in the biofilter [m^3/h];
- y = mole fraction in the gas phase;
- y^s = mole fraction in the gas phase that reaches the biomass;
- k_g = transport matter coefficient [m/h];
- a = specific surface area [m^2/m^3];
- z = direction of the oxidation (i.e. biofilter height);

- H_{CH_4} = Methane Henry's constant [dimensionless];
- H_{O_2} = Oxygen Henry's constant [dimensionless];
- H_{CO_2} = Carbon Dioxide Henry's constant [dimensionless];
- S = effective surface of the filling material in which occurs the oxidation [m^2].

For the Henry's law constants the following values were used (values expressed as gas concentration/liquid concentration and supposed constant at different temperatures):

$$H_{CH_4} = 29.2; H_{O_2} = 31.4; H_{CO_2} = 1.19 \quad [31]$$

The terms at the right hand side of the equations are related to the diffusion process that occurs within the system. Further, it can be assumed that the diffusion rate equals the reaction rate in the biofilm. The kinetics of the oxidation process are only dependent on the methane concentration at sufficiently high oxygen concentration as discussed above. Experimental data suggest that, if oxygen has a concentration value around 9%, there is the maximum oxidation rate for the oxygen and the half-saturation constant has a value that is (very) low in relation to the oxygen molar fraction ($K_m^{O_2} \ll y_{O_2}$) therefore the oxygen term can be removed from the Equation 1 [27]. As such the following equations are obtained.

$$k_g^{CH_4} a \cdot C_{TOT} (y_{CH_4} - H_{CH_4} \cdot y_{CH_4}^s) = R_{CH_4} = \frac{V_{MAX} \cdot y_{CH_4}^s}{K_m^{CH_4} + y_{CH_4}^s} \quad (5)$$

$$k_g^{O_2} a \cdot C_{TOT} (y_{O_2} - H_{O_2} \cdot y_{O_2}^s) = 2 \cdot R_{CH_4} = 2 \cdot \frac{V_{MAX} \cdot y_{CH_4}^s}{K_m^{CH_4} + y_{CH_4}^s} \quad (6)$$

$$k_g^{CO_2} a \cdot C_{TOT} (y_{CO_2} - H_{CO_2} \cdot y_{CO_2}^s) = R_{CH_4} = -\frac{V_{MAX} \cdot y_{CH_4}^s}{K_m^{CH_4} + y_{CH_4}^s} \quad (7)$$

Where C_{TOT} = total concentration of moles in the gas phase [mol/m^3];

Based on the equations above the change of the methane concentration in the biofilter height can be calculated as follows.

$$y_{CH_4}^s = \frac{y_{CH_4}}{H_{CH_4}} - \left(\frac{R_{CH_4}}{k_g^{CH_4} a \cdot C_{TOT} \cdot H_{CH_4}} \right) \quad (8)$$

$$y_{O_2}^s = \frac{y_{O_2}}{H_{O_2}} - \left(\frac{2 \cdot R_{CH_4}}{k_g^{O_2} a \cdot C_{TOT} \cdot H_{O_2}} \right) \quad (9)$$

$$y_{CO_2}^s = \frac{y_{CO_2}}{H_{CO_2}} - \left(\frac{R_{CH_4}}{k_g^{CO_2} a \cdot C_{TOT} \cdot H_{CO_2}} \right) \quad (10)$$

Dimensionless numbers

Two dimensionless numbers were defined in order to describe the behavior of the biofilter. The *Damköhler number* (Da) is a dimensionless number used to relate the reaction timescale (reaction rate) to the transport phenomena rate occurring. The *Stanton number* (St) is a dimensionless number used to relate the reaction timescale (reaction rate) to the diffusion phenomena rate occurring.

$$Da = \frac{\text{residence time}}{\text{reaction time}} = \frac{t_{RES.}}{t_{REA}} = \frac{S \cdot h}{Q} \bigg/ \frac{C_{TOT}}{R}$$

$$St = \frac{\text{residence time}}{\text{diffusion time}} = \frac{t_{RES.}}{t_{DIFF}} = \frac{S \cdot h}{Q} \bigg/ \frac{1}{k_g \cdot a}$$

The ratio between both dimensionless numbers is:

$$X = \frac{Da}{St} = \frac{R}{C \cdot k_g \cdot a}$$

Further, a new variable (ω) can be defined: $\omega = z / h$ where h is the height of the filling material inside the biofilter: the effective oxidation height. Based on the dimensionless numbers, the set of equations above (2-3-4) can be rewritten accordingly (solving Equation 8 $y_{CH_4}^s$ results in a quadratic equation and for physical reasons the negative solution was discarded).

$$\begin{aligned} \frac{dy_{CH_4}}{d\omega} &= -St_{CH_4} \cdot (y_{CH_4} - H_{CH_4} \cdot y_{CH_4}^S) \\ y_{CH_4}^S &= \frac{-\left(\frac{X_{CH_4}}{H_{CH_4}} - \frac{y_{CH_4}}{H_{CH_4}} + K_m^{CH_4}\right) + \sqrt{\left(\frac{X_{CH_4}}{H_{CH_4}} - \frac{y_{CH_4}}{H_{CH_4}} + K_m^{CH_4}\right)^2 + 4 \frac{K_m^{CH_4} y_{CH_4}}{H_{CH_4}}}}{2} \end{aligned} \quad (11)$$

$$\begin{aligned} \frac{dy_{O_2}}{d\omega} &= -St_{O_2} \cdot (y_{O_2} - H_{O_2} \cdot y_{O_2}^S) \\ y_{O_2}^S &= \frac{y_{O_2}}{H_{O_2}} - 2 \cdot \frac{X_{O_2}}{H_{O_2}} \cdot \frac{y_{CH_4}^S}{K_m^{CH_4} + y_{CH_4}^S} \end{aligned} \quad (12)$$

$$\begin{aligned} \frac{dy_{CO_2}}{d\omega} &= St_{CO_2} \cdot (y_{CO_2} - H_{CO_2} \cdot y_{CO_2}^S) \\ y_{CO_2}^S &= \frac{y_{CO_2}}{H_{CO_2}} - \frac{X_{CO_2}}{H_{CO_2}} \cdot \frac{y_{CH_4}^S}{K_m^{CH_4} + y_{CH_4}^S} \end{aligned} \quad (13)$$

Finally the degree of methane conversion (x_{CH_4}) was defined as:

$$x_{CH_4} [\%] = \left(\frac{y_{CH_4,0} - y_{CH_4}}{y_{CH_4,0}} \right) \cdot 100$$

Determination of the model's parameters

Transport coefficient k_g

The term $k_g \cdot a$ that appears in the Equations 2, 3 and 4 is the volumetric transport coefficient. Seongyup Kim et al. [32] consider the absorption of CO_2 in a biofilter filled with compost and woodchips and obtained the following function in relation to the volumetric flow rate:

$$(k_g \cdot a)_{CO_2} = 69.42 \cdot \left(\frac{Q}{S} \right)^{0.46}$$

The following functions were used for the two other variables [33], as transport coefficients can be correlated by the diffusion coefficient (D).

$$\bullet \quad (k_g \cdot a)_{CH_4} = (k_g \cdot a)_{CO_2} \cdot \sqrt{\frac{D_{CH_4}}{D_{CO_2}}}$$

$$\bullet \quad (k_g \cdot a)_{O_2} = (k_g \cdot a)_{CO_2} \cdot \sqrt{\frac{D_{O_2}}{D_{CO_2}}}$$

In water at 25°C for O_2 D is $20 \text{ cm}^2/\text{s}$, for CO_2 is $19.2 \text{ cm}^2/\text{s}$ and for CH_4 is $14.9 \text{ cm}^2/\text{s}$. Generally only the diffusion coefficient in the liquid phase (water) is known but in this case the diffusion occurs in a biofilm system. In this kind of system for light gases (e.g., O_2 , CO_2 or CH_4) the ratio between the mass transfer diffusion coefficient effective and the mass transfer diffusion coefficient in the liquid phase is 0.6 ^[34].

Michaelis–Menten kinetics

The value chosen for V_{max} was initially $8.1 \text{ mol}/(\text{m}^3 \cdot \text{h})$ ^[35] and its temperature dependence was accounted for by multiplying the maximum value of the methane oxidation rate by a correction factor $f_{V,T}$ ^[35] which was assumed to be proportional to the temperature in °C, up to a junction temperature T_j ($10/\ln 2.8 = 9.71^\circ \text{C}$).

$$f_{V,T} = 2.8^{\frac{T-22^\circ \text{C}}{10}} \quad (T > T_j)$$

$$f_{V,T} = \frac{\ln 2.8}{10} \cdot 2.8^{\frac{1}{\ln 2.8} - 2.2} \times T \quad (T < T_j)$$

The parameter K_m , was also made a function of temperature. Based on the data presented by De Visscher et al. ^[35] the half-saturation constant was expressed as a function of temperature (*Fig. 2*):

$$K_m = 0.000729 \quad \text{for } T < 5^\circ \text{C}$$

$$K_m = -1.62861 \cdot 10^{-7} T^3 + 9.26 \cdot 10^{-6} \cdot T^2 - 3.54 \cdot 10^{-6} \cdot T + 6.4 \cdot 10^{-4} \quad \text{for } 5 < T < 35^\circ \text{C}$$

$$K_m = 0.005171 \quad \text{for } T > 35^\circ \text{C}$$

Sensitivity Analysis

For the sensitivity analysis two functions were considered ^[36]. First, the *Local Sensitivity Function* (*SF*) was calculated: $SF = (V^* - V)/(P^* - P)$; where P represent the parameter value, V is the variable calculated with the original parameter value while P^* represents the perturbed parameter value, i.e. $(1 + 10^{-3}) \cdot P$, and V^* is the variable calculated with the perturbed parameter value. Then the *Relative Sensitivity Function* (*RSF*), i.e. a normalized *Sensitivity Function* to compare the values of each variable was calculated. This allows for comparing different sensitivity functions:

$$RSF = (SF \cdot P)/V$$

Results and Discussion

Overall operation

The standard size of a bio-container in which occurs the oxidation and also other operating conditions are given in *Table 1*. These values are based on actual operational values for biofilter treating biogas with a low content of methane. Based on these values, the biofilter flow regime can be determined. The Damköhler number (Da) is 0.12 while the Stanton number is 44.11 . As such, $X = Da/St = 0.12/44.11 = 0.003$. As $X \ll 1$ the reactor is in a kinetic regime.

In these conditions the degree of methane conversion is 48.3% and the model gives the results shown in *Figure 3*.

Influence of operational parameters

The importance of the operational parameters such as flow rate, methane concentration and temperature on the process of methane oxidation can be determined. Some parameters mentioned in *Table 1* where varied as indicated in *Table 2*. The effect were investigated in 3 different scenarios and presented in *Figure 4*.

The results demonstrate that the increase of the flow rate value involves a worsening of the oxidative process in terms of methane conversion. This deterioration is due to the fact that increasing the flow rate decreases the residence time of the gas inside the system. The increase of the methane concentration in the gaseous flow involves a worsening of the conversion. The methane concentration in the landfill gas flow is a non-manipulatable variable of the system, being fixed by the type of gas produced by landfill. However, considering that the methane concentration in the landfill gas tends to decrease over the years, it can be expected, on the basis of these results, an improvement in time of the performance of the biofilter.

Finally, the biological processes are strongly depending on the temperature changes that takes place in the biofilter. This is mainly due to the temperature dependence of the kinetic parameters of the biological reactions (although in reality also other variables are affected, although to a lesser extent). For this reason it is important to determine the system operation by varying the temperature value inside the biofilter. As the biofilter is integrated into the landfill cover system and is not externally heated, the temperature regime of the investigated biofilter in general follows ambient temperature, with the extent varying according to the thermal conductivity of the chosen filter materials ^[16]. From the simulations it can be seen that the degree of methane conversion is an increasing as a function of temperature. Methane oxidation reached a maximum between 25 and 30°C with a consistent increased rate from 12 to 25°C ^[37]. Whalen et al. ^[38] reported 31°C as optimum temperature for a landfill cover soil, King et al. ^[39] reported the range 19-38 °C for pure cultures of methanotrophs.

Iso-conversion graph

From an operational point of view, it is possible to draw graphs that can be particularly useful for the biofilter in the design and subsequent management phase. This graphs show the iso-conversion curves, i.e. the locations of the points characterized by having the same values of efficiency of methane degradation. Considering three different methane mole fractions (10% - 20% - 40%), two

different values of V_{MAX} ($8.1 - 4 \text{ mol/m}^3 \cdot \text{h}$) and defining the residence time of the gas in the reactor as $t_p [\text{h}] = V/Q_T$ (where V is the effective volume of the system and Q_T is the effective flow that arrives to the system). Simulation at a lower V_{MAX} aimed at demonstrating the behaviour of the biofilter under sub-optimal conditions. *Figure 5* is constructed considering a methane conversion range between 10% and 95%.

Sensitivity Analysis

Sensitivity analysis helps to understand the dynamics of the system and the contribution of the various parameters to the model output uncertainty and the system performance in general. For this sensitivity analysis the y_{CH_4} , y_{CO_2} and y_{O_2} (molar concentrations in the gas phase exit from the biofilter) were considered as variables. The parameter values mentioned in Table 1 were used. Above it was discussed that several of these parameters are related e.g. $kg \cdot a$ is based on the applied Q , and also K_m and V_{max} are functions of the temperature. For this sensitivity analysis $kg \cdot a$, K_m and V_{max} were disconnected from their dependence from flow and temperature respectively. As such, a fixed value as indicated in Table 3 was used. For each parameters and for each variables it is possible to calculate the SF and the RSF and the numerical results of this calculation, using the nominal operating conditions of *Table 1*. The resulting sensitivities are presented in *Table 3*: the cells with a light gray shading have an RSF between 0,25 and 0,8 which means that they are influential. The cells with a dark grey shading have a RSF higher than 0,8 which means that they are very influential. All other cells have a RSF lower than 0,25 which means that they are not influential ^[40].

According to *Table 3* the parameters that influence the system are the maximum oxidation rate of methane (V_{MAX}), the half-saturation constant (K_m), the Temperature (T) and the Flow (Q). This is the reason why for the scenario analysis above Q , y_{CH_4} and T were used as parameters. Others models in literature found that the system is very sensitive to V_{max} e.g. De Visscher et al. ^[27], Stein et al. ^[26].

Morris screening

In the Morris screening ^[41] the (relative) sensitivity functions of different parameters are calculated for different parameter values. This approach has been successfully applied in modelling of environmental (e.g. water) systems ^[42-43]. The two most influential biological parameters (maximum oxidation rate of the methane V_{MAX} and the half-saturation constant K_m) were used. For both parameters, three different values were used, resulting in nine different parameter combinations for which a (relative) sensitivity functions was calculated. Initially the Morris screening was performed around the values proposed in literature (V_{max} of 8.1 mol/(m³*h) and K_m of 0.003). As such V_{max} values of 7.9, 8.1 and 8.3 mol/(m³*h) and K_m values of 0.001, 0.003 and 0.005 were used. In a second screening, the V_{max} range was extended to 4, 6 and 8.1 mol/(m³*h) in order to see the effect of a broader V_{max} range. The values for K_m were left unchanged. As such in total 18 different combinations of V_{max} and K_m were used in the Morris screening and the calculation of the local sensitivity functions.

With the global results of the Morris screening (using the nominal operating conditions of Table 1) it's possible to achieve a simple statistical analysis to calculate the average and the standard deviation of these 18 calculated RSF for K_m and V_{max} (Table 4).

The values demonstrate once again that V_{max} is the most important parameter for the model. While it may be acceptable to use an V_{max} only as function of temperature, there remains the problem of determining what magnitude to use. The V_{max} parameter is a function of several variables, including soil particle-size distribution, moisture content, gas concentrations, nutrient status, and pH ^[26]. According to the work of Hilger et al. ^[25], the accumulation of exopolymeric substances (slime) produced by the methanotrophic bacteria is also an important factor controlling V_{max} . As such, careful determination of V_{max} is important.

Conclusions

A novel model was developed in this work to describe the methane oxidation in a biofilter. The model was developed for the most important biogas components (methane, oxygen and carbon dioxide) and considers several parameters such as gas transport, temperature, flow quantity, gas concentration. The sensitivity analysis has shown that the model is most sensitive to V_{max} , so this parameter need to be determined accurately.

The behaviour of the model was investigated from a theoretical point of view using the data available in literature. Specifically, it was found that temperature, flow and methane concentration are the most important parameters that influence the system. The iso-conversion curves give practical informations to design and manage a biofilter.

Future work should be focussing on developing lab-scale and field-scale biofilter in order to accurately determine the most sensitive parameter, V_{max} and extend the model beyond the current knowledge (e.g. temperature higher than 30°C and different environmental conditions). Data from such systems can be used for model testing and validation.

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FIGURE CAPTIONS

Figure 1. Cross-sectional details of the Biofilter

Figure 2. Variation law of K_m (dimensionless) as a function of temperature

Figure 3. Variation of the mole fraction as a function of the biofilter height

Figure 4. Variation of the degree of methane conversion x_{CH_4} as a function of the biofilter height by varying: a) the flow from 10 to 100 Nm³/h (scenario 1); b) the methane mole fraction from 5% to 50% (scenario 2); c) the temperature from 1 to 30°C (scenario 3).

Figure 5. Iso-conversion curves for a biofilter ($V = 22.75 \text{ m}^3$) as a function of the residence time of the landfill gas and of the temperature of the system with these conditions: a) $y_{\text{CH}_4} = 10\%$ - $V_{\text{MAX}} = 8.1 \text{ mol}/(\text{m}^3 \cdot \text{h})$ b) $y_{\text{CH}_4} = 20\%$ - $V_{\text{MAX}} = 8.1 \text{ mol}/(\text{m}^3 \cdot \text{h})$ c) $y_{\text{CH}_4} = 40\%$ - $V_{\text{MAX}} = 8.1 \text{ mol}/(\text{m}^3 \cdot \text{h})$ d) $y_{\text{CH}_4} = 10\%$ - $V_{\text{MAX}} = 4 \text{ mol}/(\text{m}^3 \cdot \text{h})$ e) $y_{\text{CH}_4} = 20\%$ - $V_{\text{MAX}} = 4 \text{ mol}/(\text{m}^3 \cdot \text{h})$ f) $y_{\text{CH}_4} = 40\%$ - $V_{\text{MAX}} = 4 \text{ mol}/(\text{m}^3 \cdot \text{h})$.

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. Operating conditions used in the simulations for scenario analysis

Parameters	Scenario 1	Scenario 2	Scenario 3	UM
Q	<i>Variable</i>	50	50	Nm ³ /h
T	30	30	<i>Variable</i>	°C
C _{TOT.}	43.8	43.8	43.8	mol/m ³
CH ₄	20	<i>Variable</i>	20	%
CO ₂	20	20	20	%
O ₂	21	21	21	%

Table 3. Values of Sensitivity Functions

	Normal Value	[CH ₄]		[CO ₂]		[O ₂]	
		SF	RSF	SF	RSF	SF	RSF
Km [dimensionless]	0.005	372.60	0.44	372.60	0.14	745.21	0.93
Vmax [mol/(m ³ *h)]	8.1	0.48	1.04	0.48	0.29	0.97	2.48
Kg*a CH ₄ [h ⁻¹]	102.55	0.00	0.01	0.00	0.00	0.00	0.01
Kg*a CO ₂ [h ⁻¹]	116.41	0.00	0.00	0.00	0.00	0.00	0.00
Kg*a O ₂ [h ⁻¹]	104.67	0.00	0.00	0.00	0.00	0.00	0.00
T [°C]	25	0.33	2.19	0.33	0.60	0.66	5.23
Q [Nm ³ /h]	50	0.08	1.04	0.08	0.29	0.16	2.49

Table 4. Average value (with standard deviation) of the calculated relative sensitivity functions calculated during the Morris screening

Variable	Vmax		Km	
	Applied Vmax values:		Applied Vmax values:	
	7.9 - 8.1 - 8.3	4 - 6 - 8.1	7.9 - 8.1-8.3	4 - 6 - 8.1
CH ₄	1.76 ± 1.04	0.4515 ± 0.1856	0.57 ± 0.14	0.0081 ± 0.0062
CO ₂	0.3 ± 0.02	0.2289 ± 0.0528	0.11 ± 0.03	0.0032 ± 0.0018
O ₂	5.12 ± 3.68	1.7872 ± 1.2555	1.6 ± 0.63	0.0250 ± 0.0224