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A mathematical steady state model for methane bioelimination in a closed biofilter

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ABSTRACT

Methane is a greenhouse gas, emitted from sources such as landfills. This paper presents a steady state model of methane biofiltration taking into consideration the impact of various parameters, such as the inlet methane concentration, the gas superficial velocity and the packing bed average temperature, on the methane biofilter efficiency. More specifically, the model developed here considers that the average bed temperature is influenced by the elimination capacity of methane in the biofilter, which is function of the methane inlet load. When using this model, it is possible to estimate the biofilter performance in terms of parameters, such as the conversion, elimination capacity and carbon dioxide production. Comparison of the model generated performance values with experimental data in the range of methane concentrations varying from 1500 to 9500 ppmv yields satisfactory results (<2–10% error, depending on the inlet methane concentration and on the performance parameter).

1. Introduction

Methane (CH_4) is a greenhouse gas that is 21 times more detrimental to atmospheric stability than carbon dioxide (CO_2) [1,2]. The emissions of this pollutant to atmosphere are mainly related to energy generation, agriculture and waste disposal landfills. Several earlier studies have demonstrated that the CH_4 emitted from these sources can be efficiently controlled by means of biofiltration [2]. Methane is assimilated by the methanotrophs: these bacteria use the CH_4 as their sole and unique carbon source to satisfy their metabolic needs. In addition to the methanotrophic population, the availability of nutrients (mainly as nitrogen and phosphorus elements) and supportive operating conditions, such as the moisture of the packing material, the gas superficial velocity and the inlet CH_4 concentration, are all to be accounted for among the factors needing to be closely controlled for operating a successful bioprocess.

During the past 3 decades, several mathematical models dealing with the biofiltration of pollutants, such as volatile organic or inorganic compounds, have been developed in order to predict the efficiency of the bioprocess and the behaviour of the pollutants, and products. Indeed, Ottengraf and Van den Oever [3] were the first authors to propose a model which has since been used as the basis for more sophisticated ones developed later by authors such as Shareefdeen et al., Hodge and Devinny or Deshusses et al. [4–7]. All the characteristics including the similarities and differences of these models can be found in the books published by Devinny et al. [8], Kennes and Veiga [9] and in many other reviews published more recently [10,11]. In general, models take into account physical, chemical and biological phenomena occurring in biofiltration such as diffusion, mass transfer of the pollutants and biodegradation kinetics, which have been described in detail in the review conducted by Devinny and Ramesh [10].

According to our knowledge, in the particular case of CH_4, it is to be noted that only one empirical model relative to its bioelimination in a compost-based bed is available in the state-of-the-art [12]. Further, some simulation models, which took into account the gas transport and the CH_4 bioelimination through a two-substrate Michaelis–Menten equation, were also elaborated [13,14]. Their main objective was to predict the performance of CH_4 biooxidation achieved in landfill-covered soil under a passive aeration regime.

The objective of the present study is to develop a simple steady state model for CH_4 biofiltration in a closed and actively aerated system that takes into account the important operating parameters, such as the inlet CH_4 concentration, the gas superficial velocity and the packed bed average temperature, affecting the CH_4 biofilter efficiency. The study is based on that of Jorio et al., 2003 which was applied to styrene [15]. The present model integrates kinetic data that has been previously obtained from a CH_4 biofilter [16] and also the biofilter average temperature, which appears to be the main novelty. Such a model could then be used to estimate the overall

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Nomenclature

A	specific surface of the packing material (m^2/m^3)
C_P	concentration of P within the gas phase (g/m^3)
D	biofilter inlet diameter (m)
D_P	diffusion coefficient of P in the biofilm (m^2/s)
EC	elimination capacity ($g/m^3/h$)
H	height of the packing material within the biofilter (m)
H_P	Henry coefficient of P (dimensionless)
IL	inlet load ($g/m^3/h$)
in	inlet flow
$k(T)$	maximum substrate utilisation rate at a T temperature (1/s)
K_m	Monod constant (g/m^3)
out	outlet flow
P	CH_4 or CO_2
P_{endo}	endogenous production of CO_2 ($g/m^3/h$)
P_{CO_2}	CO_2 production ($g/m^3/h$)
Q	gas flow rate (m^3/h)
r	CH_4 consumption rate (g/m^3 (filter bed)/s)
S_P	concentration of P within the biofilm (g/m^3)
t	time (s)
T	temperature (°C)
u_g	gas superficial velocity within the biofilter ($m^3/m^2/s$)
V	biofilter bed volume (m^3)
x	depth coordinate in the biofilm (m)
X_b	density of biomass in the biofilm (g/m^3)
X	conversion (%)
Y	biomass yield coefficient (g biomass/g CH_4)
z	biofilter height coordinate (m)

Greek symbols

α_{CO_2/CH_4}	yield of CO_2 (g gaseous CO_2 produced/g CH_4 consumed)
δ	biofilm thickness (m)
ε	filter bed porosity (dimensionless)
μ	specific growth rate of the microorganisms within the biofilm (1/s)
μ_m	maximum specific growth rate of microorganisms within the biofilm (1/s)
σ	CH_4 consumption rate (g/m^3 (filter bed)/s)
θ	temperature coefficient (dimensionless)

biofilter efficiency in terms of the conversion, elimination capacity and carbon dioxide production (P_{CO_2}).

2. Model development

To generate the mathematical model of the methane biofilter presented in this paper, some simplifying assumptions have been made and are described below.

1. In the CH_4 biofilter, there is no oxygen (O_2) supply limitation because O_2 is abundantly present (around 20% (v/v) for O_2 against < 1.2% (v/v) for CH_4) and is more readily transferred than CH_4. The diffusion coefficients are respectively 2.5×10^{-5} and 1.49×10^{-5} cm^2/s for O_2 and CH_4, at a temperature of 298 K [15,17].
2. The gas flow follows a plug-flow regime.
3. In general, models consider a planar geography (which is acceptable for biofilm because of its thinness compared to the filter bed particles' curvature) and assume that the biofilm covers the packing material particle entirely. This present model will adopt similar assumptions. It will be noted that, in some particular cases, the existence of uncovered filter material particles are considered and generally, it is supposed that the uncovered surface intervenes in the process, through adsorption, if the packing material permits it. However, in our present case, the pollutant CH_4 cannot be readily adsorbed under the present operating conditions for example, gas humidity is >80% and the present inorganic packing material is not generally used as an adsorbent [18].
4. The biofilm is uniform in the biofilter, having constant and uniform density and other characteristics, including its thickness (estimated at an average value of 85 μm).
5. The density of the biomass is constant over the entire biofilter, that is, the net growth of microorganisms in biofilm is zero. This assumption is believed to be acceptable for the steady state operation, after the start-up period is completed, since almost no clogging was seen to occur within the biofilter over a year [19].
6. The biodegradation occurs only on the external surface of each particle of filter bed material. This means that the biodegradation occurring inside the pores is neglected.
7. The process products (e.g. CO_2) do not affect the CH_4 removal micro-kinetics in the biofilter.
8. The effects of the irrigation will be neglected. It is assumed that the moisture control within the biofilter does not disturb its behaviour (excessive moisture is detrimental to the elimination of hydrophobic compounds such as CH_4 in biofilters [20]).
9. The packing material's temperature will be considered to be uniform over all of the inorganic biofilter length for each experimental condition, as revealed in previous experiments [21].
10. It will be considered that the elimination capacity (EC) is doubled after the operating temperature has been increased by 7 °C (for biofilter temperatures below 35 °C). According to the literature, a temperature increase of some 5–10 °C is necessary to double the EC (an average value of 7 °C has been adopted in this study) [22,23].
11. It will be supposed that the average temperature of the packing material only affects the values of the growth rates and the Henry coefficients.
12. The diffusion coefficients are independent of the temperature and of the biomass density. This assumption has been made because it is difficult to estimate the impact of the temperature variation on these parameters. In general, the equations used to assess diffusion coefficients in water solutions are not very precise: they usually allow 6–20% error. Therefore, experimental values determined at 25 °C have been preferred [24]. On the other hand, some authors consider that the diffusion coefficient within the biofilm of a pollutant, for example phenol and toluene, varies depending on the density of the biomass [15,25]. However, the present knowledge concerning CH_4 does not permit to assess the influence of the biomass density on the CH_4 mass transfer nor does it prove that the equation, as proposed by Fan et al. [25], can be efficiently applied to the CH_4 case.
13. Mass transfer resistance is negligible for CH_4 and CO_2 in the air. In addition, CH_4 and CO_2 concentrations at air/biofilm interface are always in equilibrium as dictated by Henry's law.
14. The mass transfer at the gas–biofilm interface which depends on several parameters, including the hydrodynamics of the gas flow and the packing characteristics, that is, the surface area and bed porosity [26] is described using Fick's law.

A summary of the general phenomena, as considered in this model, is presented in Fig. 1. On the other hand, the values of the model parameters are indicated in Table 1.

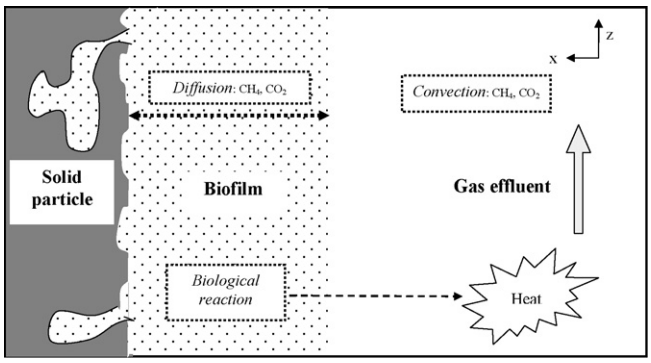

Fig. 1. Phenomena occurring within the biofilter and considered in the present model.

2.1. Mathematical model

2.1.1. In the gas phase

Two components are of particular interest in the gas phase: CH_4 and CO_2. The general mass balances within the gas phase involve essentially three terms: an accumulation term, a convection term, and a mass exchange term through the interface of the gas phase with the biofilm. These three phenomena are described by the following equation:

$$-u_g \frac{\partial C_P}{\partial z} + D_P A \left(\frac{\partial S_P}{\partial x} \right)_{x=0} = \varepsilon \frac{\partial C_P}{\partial t} \quad (1)$$

During steady state operations, there is no accumulation of the pollutant, of the by-products or of the products within the biofilter. Eq. (1) therefore becomes

$$-u_g \frac{\partial C_P}{\partial z} + D_P A \left(\frac{\partial S_P}{\partial x} \right)_{x=0} = 0 \quad (2)$$

The steady state initial condition can then be written as follows:

$$\text{At} \quad z = 0, \quad C_P(z) = C_{P,\text{in}} \quad (3)$$

2.1.2. In the biofilm

Within the biofilm, CH_4 becomes biodegraded, forming CO_2, among others. The mass balances within the biofilm are then dominated by 3 main stages: diffusion, accumulation and biodegradation. Therefore, the mass balances can be written as follows:

$$\text{Methane}: \; D_{CH_4} \frac{\partial^2 S_{CH_4}}{\partial x^2} - r = \frac{\partial S_{CH_4}}{\partial t} \quad (4)$$

$$\text{Carbon dioxide}: \; D_{CO_2} \frac{\partial^2 S_{CO_2}}{\partial x^2} + \alpha_{CO_2/CH_4} r = \frac{\partial S_{CO_2}}{\partial t} \quad (5)$$

Table 1
Values of the model parameters.

Parameter	Values	References
A	2750 m^2/m^3	–
D	0.015 m	–
D_{CH_4}	1.49×10^{-9} m^2/s	[17]
D_{CO_2}	1.96×10^{-9} m^2/s	[15]
K_m	5.37 g/m^3	[16]
P_{endo}	7 $g/m^3/h$	–
V	0.0177 m^3	–
X_b	100,000 g/m^3	–
Y	0.34 g biomass/g CH_4	[16]
H	1 m	–
α_{CO_2/CH_4}	2.01 g CO_2/g CH_4	–
ε	0.40	–
δ	85×10^{-6} m	–
μ_m	4.98×10^{-6} s^{-1}	[16]
θ	1.104	–

For a steady state operation, there is no accumulation and Eqs. (4) and (5) become, respectively,

$$\text{Methane}: \; D_{CH_4} \frac{\partial^2 S_{CH_4}}{\partial x^2} - r = 0 \quad (6)$$

$$\text{Carbon dioxide}: \; D_{CO_2} \frac{\partial^2 S_{CO_2}}{\partial x^2} + \alpha_{CO_2/CH_4} r = 0 \quad (7)$$

The steady state boundary conditions inside the biofilm can be written as follows:

$$\text{At } x = 0 \text{ and for } 0 \le z \le H: \quad S_P(0, z) = \frac{C_P(z)}{H_P(T)} \quad (8)$$

$$\text{At } x = \delta \text{ and for } 0 \le z \le H: \quad \frac{\partial S_P(\delta, z)}{\partial x} = 0 \quad (9)$$

where $H_P(T)$ depends on the temperature T (in °C) and is derived from the following equations [27]:

$$\frac{1}{H_{CH_4}} = 4.559(T + 273.15) \times 10^{(675.74/(T+273.15)-6.880)} \quad (10)$$

$$\frac{1}{H_{CO_2}} = 4.559(T + 273.15) \times 10^{(1012.40/(T+273.15)-6.606)} \quad (11)$$

2.1.3. The biodegradation

Previous experiments have shown that the Monod model can be used to describe the kinetics of the biodegradation occurring within the biofilm when the CH_4 concentration is not greater than 14,500 ppmv [16]. Therefore, one may set

$$\mu = \frac{\mu_m S_{CH_4}}{K_m + S_{CH_4}} \quad (12)$$

Hence, the CH_4 consumption rate can be written as follows:

$$r = \frac{X_b}{Y} \mu \quad (13)$$

One can define: $k(T) = \mu_m/Y$ where $k(T)$ is the temperature-dependant maximum substrate utilisation rate (1/s). Since this dependence can be modeled using the van't Hoff-Arrhenius equation [27], the following equation can be written:

$$\frac{\mu_m}{Y} = k(T) = k_{20°C} \theta^{T-20} \quad (14)$$

with $k_{20°C}$ (1/s) being the maximum substrate utilisation rate at 20 °C: $k_{20°C} = 1.464 \times 10^{-5}$ [1/s] (after experimental determination of μ_m and Y, at 20 °C) [16].

As the elimination capacity is doubled following an increase in the temperature of 7 °C (from 20 to 27 °C), one can determine that $\theta = 1.104$.

As a result, Eq. (14) becomes

$$k(T) = 1.464 \times 10^{-5} \times 1.104^{T-20} \quad (15)$$

The biodegradation term in Eq. (6) is now written as

$$r = \sigma(S_{CH_4}, T) = \frac{X_b k(T) S_{CH_4}}{K_m + S_{CH_4}} \quad (16)$$

The steady state operating temperature, T, is related to the pollutant elimination capacity of the overall process: $T = g(EC)$. This EC is directly computed from the gas pollutant concentration at the biofilter outlet, $C_{CH_4}(H)$, and thus a relation of the form $T = f(C_{CH_4}(H))$ must hold for a stationary state to be reached.

In summarizing this section, the equations describing the biofilter mathematical model may be outlined as follows: find the temperature T, the concentrations $C_{CH_4}(z)$ and $C_{CO_2}(z)$ contained in the gas phase and the concentrations $S_{CH_4}(x, z)$ and $S_{CO_2}(x, z)$ inside the biofilm, which are the solutions of the system of coupled, algebraic-differential equations:

$$T = f(C_{CH_4}(H)) \quad (17)$$

$$-u_g \frac{dC_{CH_4}}{dz} + D_{CH_4} A \left(\frac{\partial S_{CH_4}}{\partial x} \right)_{x=0} = 0, \quad 0 < z \le H, \quad C_{CH_4}(0) = C_{CH_4,in} \tag{18}$$

$$-u_g \frac{dC_{CO_2}}{dz} + D_{CO_2} A \left(\frac{\partial S_{CO_2}}{\partial x} \right)_{x=0} = 0, \quad 0 < z \le H \quad C_{CO_2}(0) = C_{CO_2,in} \tag{19}$$

$$D_{CH_4} \frac{\partial^2 S_{CH_4}}{\partial x^2} - \sigma(S_{CH_4}, T) = 0, \quad 0 < x < \delta \quad 0 < z \le H \tag{20}$$

$$S_{CH_4}(0, z) = \frac{C_{CH_4}(z)}{H_{CH_4}(T)}, \quad \frac{\partial S_{CH_4}(\delta, z)}{\partial x} = 0 \tag{21}$$

$$D_{CO_2} \frac{\partial^2 S_{CO_2}}{\partial x^2} + \alpha_{CO_2/CH_4} \sigma(S_{CH_4}, T) = 0, \quad 0 < x < \delta \quad 0 < z \le H \tag{22}$$

$$S_{CO_2}(0, z) = \frac{C_{CO_2}(z)}{H_{CO_2}(T)}, \quad \frac{\partial S_{CO_2}(\delta, z)}{\partial x} = 0 \tag{23}$$

$$\sigma(S_{CH_4}, T) = \frac{X_b k(T) S_{CH_4}}{K_m + S_{CH_4}} \tag{24}$$

Nonlinearities and couplings in the mathematical model do not make it possible to find an analytical solution. Therefore, a numerical procedure has been developed to approximate the biofilter state. This procedure makes use of the finite difference method for the treatment of Eqs. (20) and (22) and solves Eqs. (18) and (19) by means of a Runge–Kutta scheme. The nonlinearity resulting from temperature dependent coefficients has been taken into account by a fixed point loop (Appendix 1).

2.2. Materials and methods

2.2.1. Design of the biofiltration system

The biofilter, as modeled in this paper, is a cylindrical tube (15 cm in internal diameter), divided into 3 sections of equal size. Each section of the biofilter is packed with packing material of height 33 cm, leading to a total bed volume of nearly 18 L. The packing material used in this biofilter is of an inorganic nature (cylindrical rocks particles). According to our previous considered assumptions, only the external surface of the packing material is involved in the biodegradation process occurring within our biofilter and the surface area used for the modeling has been estimated to be around 2750 m^2/m^3.

The polluted gas, introduced at the base of the biofilter, is a mixture of humidified air and pure methane gas. The gas superficial velocities (GSV) always vary between 3.4 and 25.6 $m^3/m^2/h$ while the CH_4 concentrations range between 1500 and 9500 ppmv. On the other hand, the irrigation of the packing material within the biofilter is performed once a day, using a 1.5 L of nutrient solution, which contains almost all of the necessary nutrients, nitrogen: 0.75 g/L, phosphorus: 0.3 g/L, potassium: 0.076 g/L and micronutrients, like metallic species in traces, needed to sustain the bacterial growth. Its composition is presented elsewhere [28].

2.2.2. Description of the assessment parameters

In order to quantify the amount of the CH_4 pollutant introduced into the biofilter, the inlet load (IL) parameter will be used. This parameter is measured in g/m^3/h and is determined as the following:

$$IL = \frac{Q}{V} C_{(CH_4)in} \tag{25}$$

For the assessment of the biofilter performance, the conversion (X), expressed in %, and the elimination capacity (EC), in g/m^3/h, have both been used, according to Eqs. (26) and (27), respectively:

$$X = \frac{C_{(CH_4)in} - C_{(CH_4)out}}{C_{(CH_4)in}} 100 \tag{26}$$

$$EC = IL \frac{X}{100} \tag{27}$$

In order to obtain the predicted carbon dioxide production (P_{CO_2}), the present equation has been used:

$$P_{CO_2} = \frac{Q}{V}(C_{CO_2}(H) - C_{CO_2,in}) + P_{endo} \tag{28}$$

3. Results

3.1. Parameters' estimation

3.1.1. Temperature

In the present model, the variation in the microbial kinetic parameters with temperature change within the packing material is considered for the steady state operations (Eq. (14)). Increasing the packing material's temperature to between 25 and 35 °C is generally favourable to the biological process because of the improvement in the kinetic parameters. It is to be noted that the temperature of the packing material not only affects the microkinetics of the bioreaction, but also has an influence on the solubility of the pollutants within the biofilm since the Henry coefficients are temperature-dependant (Eqs. (10) and (11)). As a consequence, there is a relationship between the biofilter bed temperature and the CH_4 elimination capacity within the same biofilter. Simultaneously, the bioelimination of the CH_4 being an exothermic process, the higher the elimination capacity, the higher the amount of the energy transferred to the packing material, which thus increases its temperature.

Fig. 2 represents the elimination capacity as a function of the average temperature within the biofilter. It reveals that, as the EC increases in the biofilter, the average temperature of the biofilter packing material also increases, by following a linear equation (Eq. (29)):

$$EC = 15.8T - 401.9 \tag{29}$$

3.1.2. Carbon dioxide production

The CH_4 pollutant is utilized by the microorganisms to satisfy their needs in terms of both energy and carbon. Part of the pollutant will be devoted to the sustaining of the multiplication of the microorganisms while the remaining of it will be converted into the form of CO_2. The yield α_{CO_2/CH_4} (required to solve Eq. (7), under

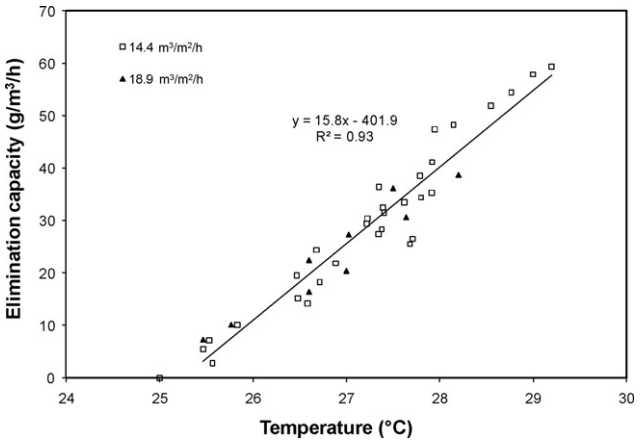

Fig. 2. Elimination capacity (g/m^3/h) expressed as a function of the average biofilter temperature (°C) for gas superficial velocities of 14.4 and 18.9 $m^3/m^2/h$.

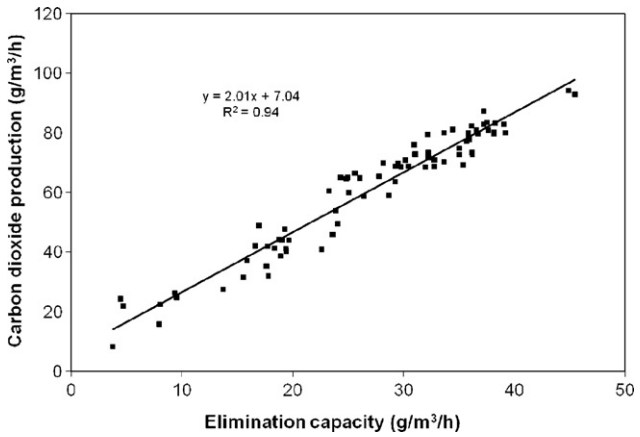

Fig. 3. Production of CO_2 (g/m^3/h), in the gas phase, as a function of the elimination capacity (g/m^3/h) of the biofilter, during steady state operation (gas superficial velocity = 14.4 m^3/m^2/h; CH_4 concentration: ≤9500 ppmv).

steady state condition) can be calculated by measuring the amount of consumed CH_4 and produced CO_2 and correcting the amount of CO_2 by the CO_2 produced due to endogenous activity.

Fig. 3 depicts the production of CO_2 in the gas phase as a function of the elimination capacity of the biofilter, during its steady state operation, at a GSV of 14.4 m^3/m^2/h and at CH_4 concentrations ranging between 1500 and 9500 ppmv. The slope of the curve corresponds to the value of α_{CO_2/CH_4}, and turns out to be 2.01; this implies that, for 1 g of the CH_4 biodegraded, some 2.01 g of CO_2 will exit from the biofilter through the gas phase. Fig. 3 also provides with the value of P_{endo} – intercept of the linear regression line – which turns to be 7 g/m^3/h.

3.2. Conversion and elimination capacity

Fig. 4a and b present the CH_4 conversion (predicted and experimental values) expressed as a function of the GSV for 2 different inlet CH_4 concentrations, that is, 7500 and 2500 ppmv. It is to be noted that these 2 values of CH_4 concentrations have been selected with the aim of representing each range of CH_4 concentration: from 1500 to 4500 ppmv (2500 ppmv) and from 4500 to 9500 ppmv (7500 ppmv). From these figures, it is observed that, as the GSV increases at a constant CH_4 concentration, the conversion decreases accordingly.

At 7500 ppmv, it can be noted that the present model offers a good estimation of the experimental data. For example, at GSVs of 3.4 and 18.9 m^3/m^2/h, the model predicts conversions of some 90.9% (92.7% for the experimental result) and 36.7% (36.0% for the experimental result). More generally, in Fig. 4a, the differences between the experimental conversions and the predicted values never exceed 2%. Also, in accordance with the model, the highest GSV, allowing CH_4 conversions ≥85%, is 4.5 m^3/m^2/h, while for a CH_4 conversion ≥95%, the GSV must be ≤2.7 m^3/m^2/h—recalling that the 85% and the 95% levels are the minimum conversions usually aimed at industry for the control of pollutants difficult to biodegrade and for the volatile organic compounds, respectively. On the other hand, at low CH_4 concentrations, that is, 2500 ppmv (Fig. 4b), based on the experimental data, GSVs of 5.8 and 4.1 m^3/m^2/h are necessary to obtain conversions ≥85% and ≥95%, respectively. The latter values are higher than those found at 7500 ppmv and confirm the point that it is easier to obtain higher conversions in the biofilter operating at lower CH_4 concentrations than in the case of biofiltration at higher CH_4 concentrations.

In addition, at 2500 ppmv, it can be observed that both the experimental and the predicted CH_4 conversions decrease with the CH_4 concentration increase at a given GSV comprised of between 3.4 and 18.9 m^3/m^2/h but the difference between the experimental and the predicted conversion values reaches up to 10%. For instance, at GSV values of 3.4 and 18.9 m^3/m^2/h, the model predicts conversions of 90% (around 100% for the experimental result) and 35% (41% for the experimental result). To explain this behaviour it can be recalled that the determination of the kinetic parameters has been effected using solid extracts from a biofilter operating at around 7500 ppmv, which were thereafter exposed to CH_4 concentrations of 2500 ppmv without any additional acclimatization provided in the biofilter [16]. This lack of acclimatization could lead to an error in the values of the kinetic parameters, which could explain why they are slightly less representative of the steady state operations at 2500 ppmv than they were at around 7500 ppmv. In addition, the Monod parameters used in this model were slightly under-evaluating the real specific growth rate (error of 2–5%) when the CH_4 concentration is ≤3000 ppmv), as presented in Delhoménie et al. [16]. The cumulating effect of those two could therefore explain the lower precision in the predicted conversion values, at 2500 ppmv.

As an overall conclusion, it is to be noted that the model developed in this study appears to be very appropriate for the CH_4 concentrations ranging from 4500 to 9500 ppmv, the difference between the experimental and estimated conversions, being ≤5% for these CH_4 concentrations (data not shown). However, the model becomes less appropriate, but still acceptable, as the CH_4 concen-

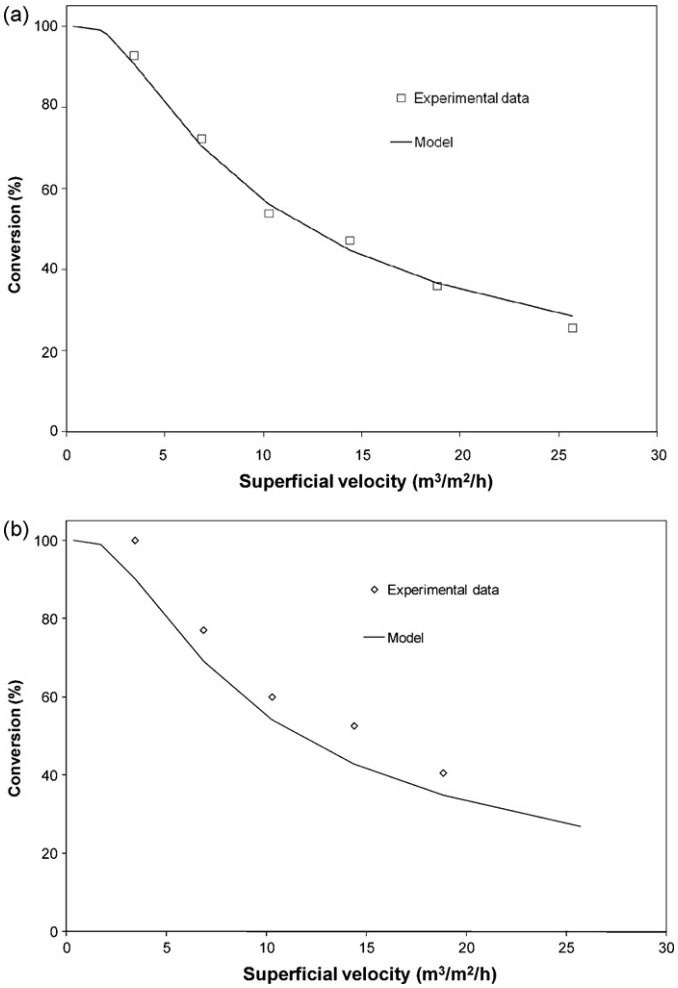

Fig. 4. (a) CH_4 conversion (predicted and experimental values) (%) expressed as a function of the gas superficial velocity (m^3/m^2/h), for an inlet CH_4 concentration of 7500 ppmv. (b): CH_4 conversion (predicted and experimental values) (%) expressed as a function of the gas superficial velocity (m^3/m^2/h), for an inlet CH_4 concentration of 2500 ppmv.

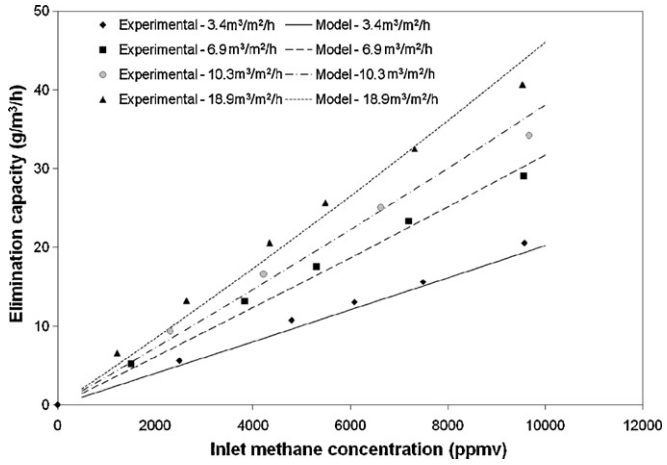

Fig. 5. Elimination capacity in the CH_4 biofilter as a function of the inlet CH_4 concentration (1500–9500 ppmv) for gas superficial velocities comprised between 3.4 and 18.9 $m^3/m^2/h$.

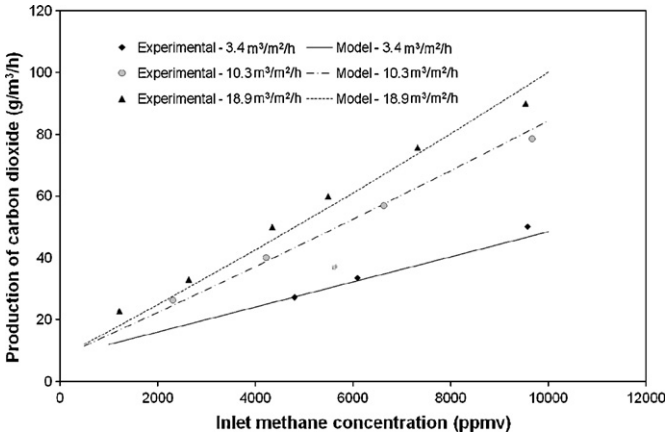

Fig. 6. CO_2 production ($g/m^3/h$) as a function of the inlet CH_4 concentration (≤9500 ppmv) for gas superficial velocities comprised between 3.4 and 18.9 $m^3/m^2/h$.

tration decreases to between 4500 and 1500 ppmv. To improve the modeling at the low CH_4 concentrations, it is important to improve the precision of the kinetic parameters involved.

Fig. 5 presents the EC obtained in the CH_4 biofilter as a function of the inlet CH_4 concentration (at around 1500–9500 ppmv). The error range noted when considering the conversion parameter for low CH_4 concentrations, for example 2500 ppmv, seems high in comparison to the EC parameter: for example, at 3.4 and 10.3 $m^3/m^2/h$ and 2500 ppmv of CH_4 concentration, the experimental EC values are 5.6 and 9.9 $g/m^3/h$ – being conversions of 100% and 60%, respectively while the predicted ones are 5.0 and 9.0 $g/m^3/h$ – being conversions of 90% and 54%, respectively. This is due to the fact that, for the lower CH_4 concentrations applied to the biofilter, the EC and IL values are low. In this case, differences between the predicted and the experimental EC that are normally minor (e.g. when the CH_4 concentration is 2500 ppmv, $\Delta EC = 0.6$ and 0.9 $g/m^3/h$ at 1 and 3 L/min, respectively), corresponds, to a high percentage difference (10% and 6%, respectively).

On the other hand, it can be observed that the EC rises with the increases in the inlet CH_4 concentration: at 6.9 $m^3/m^2/h$, after increasing the CH_4 concentration from 1500 to 9500 ppmv, the EC is increased from around 5 to around 29 $g/m^3/h$ (experimental data), or from around 5 to around 30 $g/m^3/h$ (model data). The continuous and linear increase of the EC with the inlet CH_4 concentration (Fig. 5) proves that the operating regime is diffusion-limited in the CH_4 concentrations range studied. Further, according to the model, the maximum EC that can be reached in the biofilter at a CH_4 concentration of 9500 ppmv and a GSV of 18.9 $m^3/m^2/h$, is 43 $g/m^3/h$, that is, nearly 5% higher than the experimental EC value that is around 41 $g/m^3/h$.

3.3. Production of carbon dioxide

Fig. 6 presents the P_{CO_2} as a function of the inlet CH_4 concentration, expressed in ppmv. Three GSVs have been considered in this figure: 3.4, 10.3 and 18.9 $m^3/m^2/h$. It can be observed that the increase in the P_{CO_2}, caused by the increase in the CH_4 concentration, is well predicted by the model. For example, at 10.3 $m^3/m^2/h$ of GSV, the model predicts an increase in the P_{CO_2}, from 25 to 80 $g/m^3/h$, when the CH_4 concentration is increased, from 2300 to 9500 ppmv. Accordingly, the experimental data values are 26 and 79 $g/m^3/h$ at 2300 and 9500 ppmv, respectively.

According to the present model, P_{CO_2} follows a linear trend with the inlet CH_4 concentration, for each GSV. Also, one notes that even at low CH_4 concentrations (1500–4500 ppmv), P_{CO_2} is well correlated by the model. This can be explained by the fact that the model takes into consideration the endogenous respiration parameter, P_{endo}. Indeed, the quantity of CO_2 related to the endogenous respiration represents a significant proportion (15–50%) of the total CO_2 generation occurring within the biofilter, when the EC is low.

3.4. Temperature

Fig. 7 presents the average temperature (°C) of the 3 stages of the biofilter as a function of the GSV for 4 CH_4 concentrations. For example, at 9500 ppmv value of the inlet CH_4 concentration, the experimental temperatures are 27.7 and 28.1 °C at 14.4 and 18.9 $m^3/m^2/h$, respectively. At the same time, the model uses the values of 28.0 and 28.1 °C, respectively. It is to be noted that the GSV variation (over the present study range) does not really affect the average biofilter temperature. On the other hand, when the CH_4 concentration varies, it affects the biofilter temperature.

In the present range of CH_4 concentrations, the temperature variation never exceeds 3 °C. The level of variation of the temperature must eventually be taken into consideration because an increase in temperature will favour the drying of the packing material. This latter circumstance will thereafter negatively affect the biofilter performance. To control this phenomenon and to avoid filter bed drying, the biofilter will require more frequent irrigation.

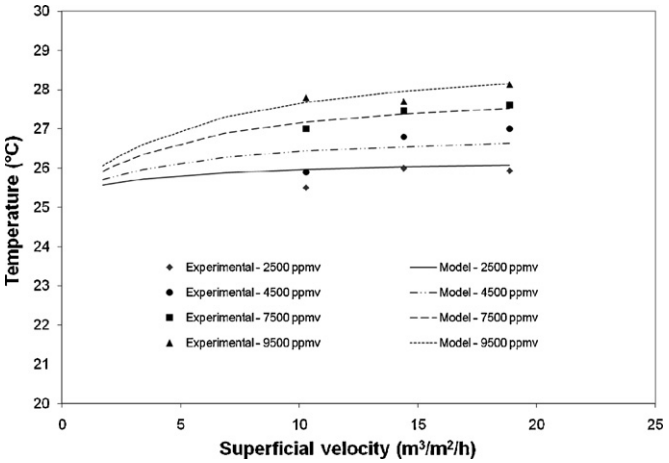

Fig. 7. Biofilter average temperature (°C) (experimental and model values) as a function of the gas superficial velocity (≤18.9 $m^3/m^2/h$).

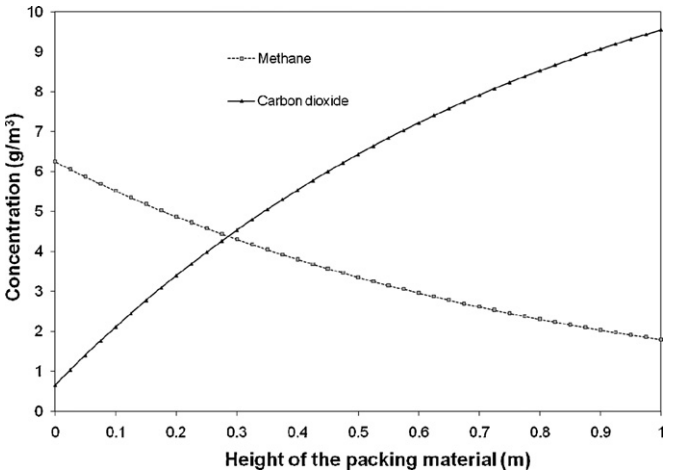

Fig. 8. Predicted concentrations of CH_4 and CO_2 within the gas phase as a function of the filter bed height (m) (inlet CH_4 concentration = 6.3 g/m^3 (or 9500 ppmv); gas superficial velocity = 6.9 m^3/m^2/h).

3.5. Profiles of concentrations in the biofilter

Fig. 8 presents the profiles of the predicted CH_4 and CO_2 concentrations in the gas phase ($C_{CH_4}(H)$ and $C_{CO_2}(H)$, respectively), as a function of the height of the filter bed within the biofilter. The inlet CH_4 concentration and the GSV in the biofilter were 9500 ppmv and 6.9 m^3/m^2/h, respectively. The profiles observed are similar to those generally observed in biofilters [29,30]. Indeed, the profiles are not linear, which confirm that the pollutant is not uniformly removed within the biofilter. For instance, at the biofilter entrance, the CH_4 and CO_2 concentrations are of 6.3 and of 0.7 g/m^3, respectively. Thereafter, at the levels z of 0.3, 0.6 and 0.9 m within the biofilter, and according to the model, the values of $C_{CH_4}(z)$ become 4.3, 3.0 and 2.1 g/m^3, while the values of $C_{CO_2}(z)$ are 4.5, 7.2 and 9.1 g/m^3, respectively. Therefore, it can be noted that the areas near the biofilter entrance are more active than the one near the biofilter exit.

4. Discussion—model's limits

In order to generate the model, the kinetic parameters that have been chosen are valid (i.e. error $\leq$5%), for CH_4 concentrations $\leq$14,500 ppmv. Above this CH_4 concentration value, other kinetic parameters must be used [16]. In addition, this model did not integrate the influence of the concentrations of nutrients, mainly nitrogen and phosphorus, in either the biofilter or in the nutrient solution. Previous experiments have demonstrated that these nutrients have a great influence on biofilter performance [28]. To apply this particular model to another biofilter, in which the nutrient concentrations are different from those in the present study, it is important to determine the kinetic parameters applicable to the new experimental nutrients' concentrations.

Also, this model considers that the biomass density, present in the biofilter, is constant. However, it has been observed that, in CH_4 biofilters, the number of living cells and their biomass density could vary, depending on the operating conditions, such as the CH_4 concentration and the biofilter history, for example the age of the biofilm [2]. The density of the biomass can affect the biofilter performance directly, through the activity of the microorganisms, and indirectly through the mass transfer. On the other hand, the influence of the biomass on the diffusion coefficient (gas–biofilm) has been neglected and the biofilm has been assimilated to a water-based phase.

5. Conclusion

The goal of this study has been to develop a model able to predict the conversion, elimination capacity, production of carbon dioxide and the packing material temperature of a closed biofilter, used to treat methane effluents. In addition to the inlet CH_4 concentration, the biofilter temperature has been considered for the model development because of its influence on the bioreaction microkinetics and on the Henry coefficients. The results obtained with this model have shown that it is appropriate for the modeling of the CH_4 conversion (i.e. with <5% difference) in biofilters used for the treatment of gas effluent with a CH_4 concentration comprised of between 4500 and 9500 ppmv in a range of GSV varying between 3.4 and 25.6 m^3/m^2/h. For lower CH_4 concentrations (<4500 ppmv), the model tends to predict conversion values lower than the experimental results (5–10% difference). On the other hand, the elimination capacity, the CO_2 production and the biofilter average temperature are well correlated over the entire range of CH_4 concentrations of interest, i.e. from 1500 to 9500 ppmv.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.cej.2009.01.032.

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