

METHANE OXIDATION IN HOMOGENOUS SOIL COVERS OF LANDFILLS: A FINITE  
– ELEMENT ANALYSIS OF THE INFLUENCE OF GAS DIFFUSION COEFFICIENT\*

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Accepted January 17, 2000

**A b s t r a c t.** Methane produced in municipal landfills covered with a soil layer diffuses to the atmosphere. Counterdiffusion of oxygen down the soil creates an oxic zone where biochemical oxidation of methane by methanotrophic microorganisms can take place. Oxidation is possible only in that part of the oxic zone where both the substrates, i.e., methane and oxygen are present simultaneously.

Biochemical oxidation of methane is governed by Michaelis-Menten kinetics. The solution of the equation system comprising the Michaelis-Menten equation generalized for the two substrates, i.e., methane and oxygen and the Fick's diffusion equation was found numerically with the use of the finite element method.

Results of the calculations were presented for the steady - state conditions in a homogenous soil layer with constant (in space and time) gas diffusion coefficient and kinetic parameters, i.e., the Michaelis constant ( $K_M$ ) and the maximum oxidation rate. The effect of the gas diffusion coefficient on the distribution profiles of methane, oxygen and actual methanotrophic activity, as well as on the depth of oxygenation zone and on the methane oxidation capacity in the soil layer is discussed.

**K e y w o r d s:** landfills, methanotrophic capacity, methane oxidation, modelling, gas diffusivity

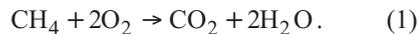
INTRODUCTION

Landfills of municipal wastes are an important source of methane and contribute significantly (6-20%) to the total anthropogenic methane emission to the atmosphere [5]. One of the possibilities to reduce the increase rate of methane concentration in the atmosphere is to oxidize the methane generated in the municipal

landfills in the soil layer used for its rehabilitation. Such possibilities have been suggested [1,2,6] and documented in the pertinent literature [4,7]. The aim of the present paper was to model the effect of gas diffusion coefficient on the efficiency of methane oxidation in a homogenous soil layer covering the landfill.

THEORY

Methane produced in the landfill can undergo biochemical oxidation in the oxic zone of the soil cover due to the methanotrophic activity of microorganisms. The reaction of oxidation can be written as follows:



The gas flux through the soil can be described by the first Fick's law:

$$F = -D \frac{dC}{dx}, \quad (2)$$

with the following meaning of the symbols:  $F$  - diffusion rate of the gas under consideration ( $\text{m}^2 \text{s}^{-1}$ ),  $D$  - gas diffusion coefficient in the soil ( $\text{m}^2 \text{s}^{-1}$ ),  $C$  - gas concentration in soil air ( $\text{m}^3 \text{m}^{-3}$ ),  $x$  - depth (m).

The change of the gas concentration with time is described by the second Fick's equation:

\*This work was supported by the State Committee for Scientific Research, Poland under grant No. 906/T09/96/10.

$$E_g \frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} \pm q \quad (3)$$

where:  $E_g$  - air-filled porosity ( $\text{m}^3 \text{m}^{-3}$ ),  $t$  - time (s),  $q$  - gas production (+) or absorption (-) rate ( $\text{m}^3 \text{m}^{-3} \text{s}^{-1}$ ).

The rate of methane oxidation is related to the concentration of the substrates, i.e., of methane and oxygen by the Michaelis-Menten equation as follows:

$$q = q_{\max} \frac{1}{\left(1 + \frac{K_{M1}}{C_1}\right) \left(1 + \frac{K_{M2}}{C_2}\right)} \quad (4)$$

where:  $q$  - actual rate of the reaction ( $\text{m}^3 \text{m}^{-3} \text{s}^{-1}$ ),  $q_{\max}$  - maximum rate of the reaction ( $\text{m}^3 \text{m}^{-3} \text{s}^{-1}$ ),  $C_1, C_2$  - concentrations of methane and oxygen, respectively ( $\text{m}^3 \text{m}^{-3}$ ),  $K_{M1}, K_{M2}$  - Michaelis-Menten constants for methane and oxygen, respectively ( $\text{m}^3 \text{m}^{-3}$ ).

Let us consider the situation of a homogeneous soil layer with constant values of diffusion coefficients for methane, oxygen, and carbon dioxide in time and in depth. In case of the steady state conditions, i.e., when the concentration of the reacting substrates and products is constant in time, as it has been assumed in our case, the Eq. (3) for methane diffusion up the soil layer is reduced to the following:

$$0 = D_1 \frac{d^2 C_1}{dx^2} - q_1. \quad (5)$$

The respective equations for the downward oxygen diffusion and for the upward diffusion of carbon dioxide are as follows:

$$0 = D_2 \frac{d^2 C_2}{dx^2} - q_2 \quad (6)$$

and

$$0 = D_3 \frac{d^2 C_3}{dx^2} + q_3. \quad (7)$$

In the situation where methane is not used by the microorganisms as the carbon source, the stoichiometric coefficients from the Eq. (1) are valid, i.e.:

$$q_2 = 2q_1 = 2q_3. \quad (8)$$

We are interested in finding the amount,  $Q$ , of methane oxidized during its diffusion from the landfill through the soil layer of depth  $L$  to the atmosphere, i.e.:

$$Q = \int_0^L q(x) dx. \quad (9)$$

As the analytical solution of the above equations is not possible because of the strong non-linearity of the term involved in the Michaelis-Menten equation, the value of  $q$  from Eq. (5) was calculated by a numerical procedure.

### Numerical procedure

To find a numerical solution of the equation system including Eqs (4)-(7) we assumed a stoichiometric reaction as described in Eq. (1), i.e., we assumed the relations:

$$q_2 = 2q_1 = 2q_3. \quad (10)$$

We decided to use the finite element method. We considered the soil layer as a set of  $n$  cells of the same dimensions, as shown in Fig. 1, and we moreover assumed that the concentrations of gases within each individual cell are constant. In this case the Eqs (5)-(7) take the form of:

$$C_1(i) = \frac{1}{2} C_1(i+1) + \frac{1}{2} C_1(i-1) - \frac{1}{2} \frac{\Delta x}{D_1} q(i), \quad (11)$$

$$C_2(i) = \frac{1}{2} C_2(i+1) + \frac{1}{2} C_2(i-1) - \frac{\Delta x}{D_2} q(i), \quad (12)$$

$$C_3(i) = \frac{1}{2} C_3(i+1) + \frac{1}{2} C_3(i-1) - \frac{1}{2} \frac{\Delta x}{D_3} q(i), \quad (13)$$

where:  $C_1(i), C_2(i), C_3(i)$  - concentrations of methane, oxygen and carbon dioxide in the  $i$ -th cell;  $q(i)$  - rate of reaction in the  $i$ -th cell;  $\Delta x$  - thickness of one cell.

We present a scheme for getting the equation of the methane concentration.

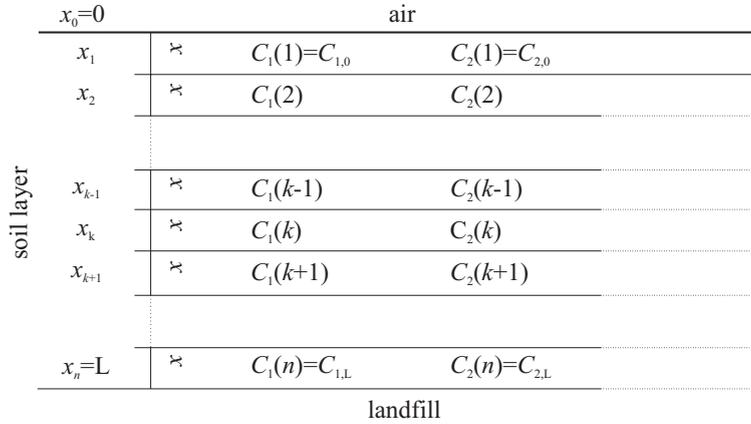


Fig. 1. Scheme of soil layers in the finite element system.

First, we rewrote the Eq. (5) in an integral form:

$$\frac{dC_1}{dx}(s_2) - \frac{dC_1}{dx}(s_1) = \frac{1}{D_1} \int_{s_1}^{s_2} q dx. \quad (14)$$

We considered the soil layer as an interval  $[0, L]$ . Then we divided it into  $n$  equal cells by putting points

$$0 = x_0 < x_1 < \dots < x_i = \frac{i}{n} L < \dots < x_n = L.$$

We approached the function  $C_1$  by a piecewise linear function  $C_1^*$  of the following form:

$$C_1^*(x) = C_1(i-1) \frac{x_i - x}{x_i - x_{i-1}} + C_1(i) \frac{x - x_{i-1}}{x_i - x_{i-1}}, \quad (15)$$

for  $x_{i-1} \leq x \leq x_i$ .

Simple differentiation of  $C_1^*$  let us to:

$$\frac{dC_1^*}{dx} = \frac{C(i) - C(i-1)}{\Delta x}, \quad (16)$$

where:  $\Delta x = x_i - x_{i-1}$ .

Putting  $s_1 = x_i, s_2 = x_{i+1}$  in the Eq. (14) for function  $C_1^*$  gave us following equation:

$$\frac{1}{\Delta x} (C_1(i+1) - C_1(i)) - \frac{1}{\Delta x} (C_1(i) - C_1(i-1)) = \frac{1}{D_1} \int_{x_i}^{x_{i+1}} q dx. \quad (17)$$

But  $q$  was constant inside the  $i$ -th cell, so we arrived at:

$$\frac{1}{\Delta x} (C_1(i+1) + C_1(i-1) - 2C_1(i)) = \frac{1}{D_1} q(i) \Delta x, \quad (18)$$

which implied the Eq. (11).

The Eqs (12)-(13) for oxygen and carbon dioxide was obtained in the same way as the above.

After computing distribution of  $q$  the integral Eq. (10) can be obtained. This method was applied for different values of the coefficients  $D, K_{M1}, K_{M2}, q_{max}$  and the thickness  $L$  of the soil layer.

The calculations were performed for the following boundary conditions: soil layer depth  $L = 1.0$  m, potential methanotrophic activity  $q_{max}$  was assumed to be  $10^{-5} \text{ s}^{-1}$ , i.e., half of the maximum value reported in literature for the landfill soil [4], the  $K_M$  value for oxygen and methane was taken as  $0.02 \text{ m}^3 \text{ m}^{-3}$ , that is the average value reported for methane in the landfill soil [4]. Methane concentration on the soil surface was assumed to be zero and at the bottom of the soil layer - as  $0.50 \text{ m}^3 \text{ m}^{-3}$ . Concentration of oxygen on the soil surface was taken as  $0.20 \text{ m}^3 \text{ m}^{-3}$  and at the soil - waste interface ( $x = L = 1.0$  m) it was taken as zero.

Diffusion coefficient of methane in the atmospheric air was assumed to be  $2.23 \cdot 10^{-5} \text{ m}^2 \text{ s}^{-1}$  while that of oxygen was taken as  $2.01 \cdot 10^{-5} \text{ m}^2 \text{ s}^{-1}$  [3].

#### RESULTS

Distribution of methane concentration for a range of relative gas diffusion coefficients from 0.005 to 0.2 is presented in Fig. 2. As it can be noted there is only a small differentiation of the oxygen distribution curves due to a large (20 fold) variation in gas diffusion coefficient. This differentiation is the most pronounced at the depth of 0.15-0.20 m and reaches 5% of oxygen concentration. Below this level, methane concentration increases linearly with depth to the level assumed for the waste layer.

Concentration of oxygen (Fig. 3) decreases curvilinearly with the depth reaching values close to zero in the depth interval from 0.2 to 0.4 m. Oxygen penetration depth increases with the gas diffusion coefficient.

Distribution curves of the methanotrophic activity  $q$  as a function of depth is shown in Fig. 4. As it could be expected, this real oxidation activity of methanotrophic microorganisms exists in the layer, where both the substrates, i.e., oxygen and methane are present simultaneo-

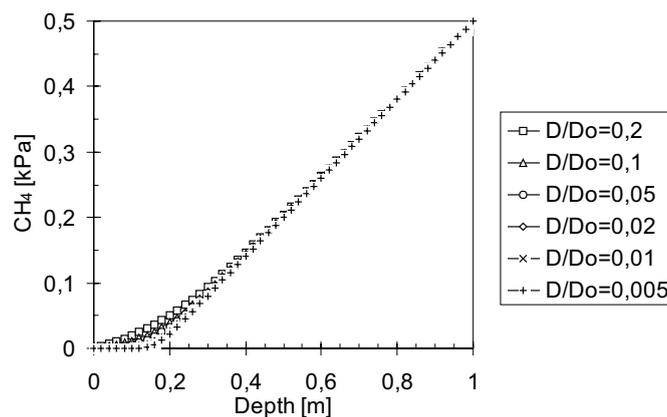
usly. The activity shows the maximum at the depths of 0.15-0.20 cm and increases with  $D/D_0$  value. At  $D/D_0 = 0.2$  the actual  $q$  value reaches in its peak 60% of the  $q_{max}$ .

Cumulative consumption of methane  $Q$  in the soil layer from  $x$  to  $L$  at different  $D/D_0$  values is presented in Fig. 5. It is evident that cumulative methane consumption in the soil layer or its methanotrophic capacity increases with the value of the relative gas diffusion coefficient.

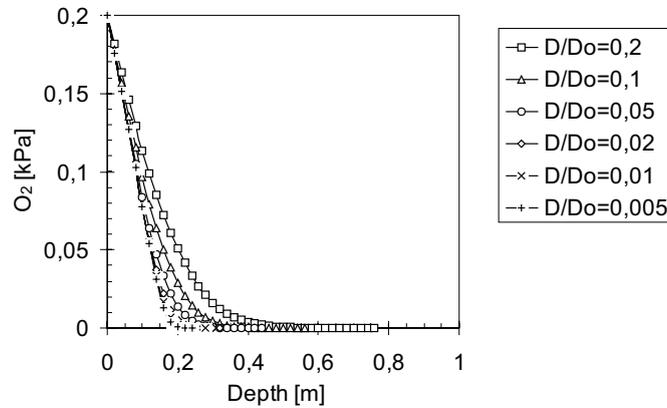
Distribution of methane flux  $F$  within the soil layer at different  $D/D_0$  values is presented in Fig. 6. It can be seen that the methane flux under biologically active layer is directly related to the gas diffusion coefficient, and that it is subsequently reduced in the methanotrophic top layer of the soil.

The ratio of methane consumption  $Q$  in the layer from  $x$  to  $L$  to the initial methane flux at the bottom of the soil layer  $F_{x=L}$  (shown in Fig. 7) decreases with the gas diffusion coefficient, and, thus, with the total methane flux, from 100% at  $D/D_0 < 0.02$  to about 70% at  $D/D_0 = 0.2$ .

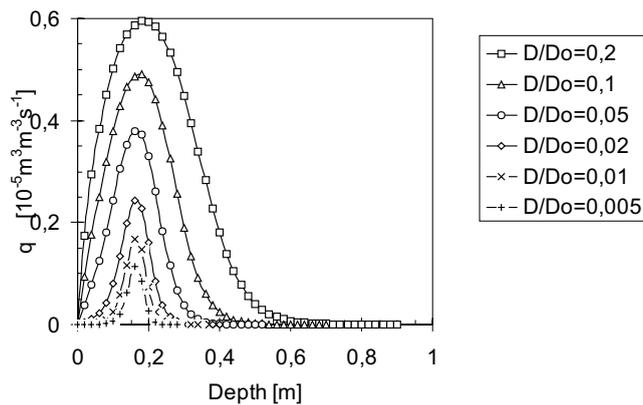
Comparison of the distribution curves of all the above mentioned parameters, i.e., of oxygen concentration, methane concentration, methanotrophic activity, methanotrophic capacity  $Q$  and the flux intensity  $F$  is presented in Figs 8 and 9 for the relative gas diffusivities 0.05 and



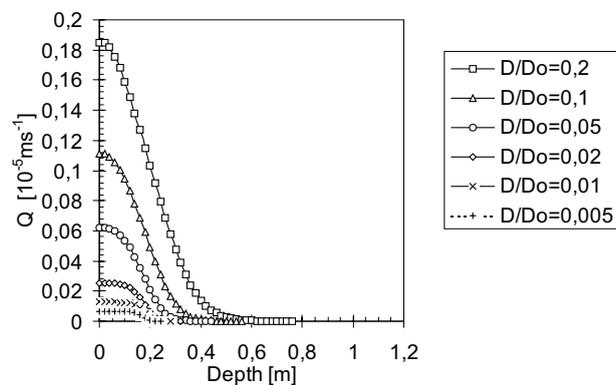
**Fig. 2.** Predicted distribution of methane concentration levels in a homogenous soil layer covering landfill at different values of relative gas diffusion coefficient  $D/D_0$  ( $D$  - gas diffusion coefficient in soil,  $D_0$  - diffusion coefficient of the same gas in the atmospheric air at the same pressure and temperature conditions; for methane at  $20^\circ \text{C}$  we assumed  $D_{0,CH_4} = 2.23 \cdot 10^{-5} \text{ m}^2 \text{ s}^{-1}$ ). Boundary conditions:  $q_{max} = 10^{-5} \text{ m}^3 \text{ m}^{-3} \text{ s}^{-1}$ ,  $K_{M_1} = K_{M_2} = 0.02 \text{ m}^3 \text{ m}^{-3}$ , total depth of soil layer  $L = 1 \text{ m}$ .  $C_{1,0} = 0$ ,  $C_{1,L} = 0.50 \text{ m}^3 \text{ m}^{-3}$ ,  $C_{2,0} = 0.20 \text{ m}^3 \text{ m}^{-3}$ ,  $C_{2,L} = 0$ .



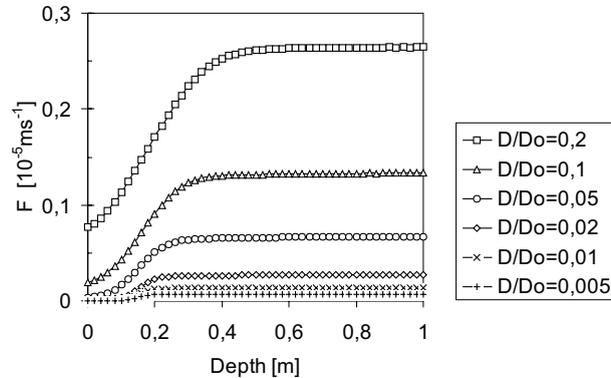
**Fig. 3.** Predicted distribution of oxygen concentration levels in a homogenous soil layer covering landfill at different values of relative gas diffusion coefficient  $D/D_0$  ( $D$  - gas diffusion coefficient in soil,  $D_0$  - diffusion coefficient of the same gas in the atmospheric air at the same pressure and temperature conditions; for oxygen at  $20^\circ\text{C}$   $D_{0,\text{O}_2}=2.01 \cdot 10^{-5} \text{m}^2\text{s}^{-1}$ ). Boundary conditions as in Fig. 2.



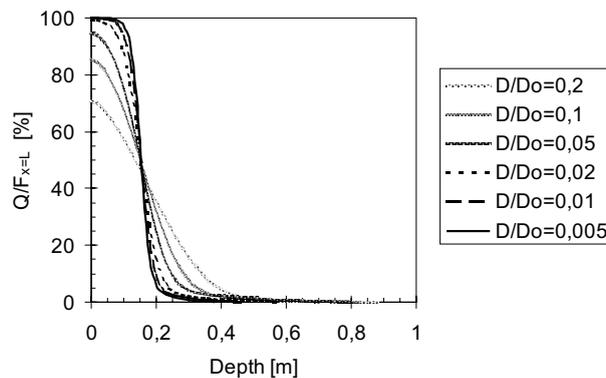
**Fig. 4.** Predicted distribution of methanotrophic activity  $q$  at different  $D/D_0$  values. Boundary conditions as in Fig. 2.



**Fig. 5.** Predicted methane consumption  $Q$  (cf. Eq. (9)) in the soil layer from  $x$  to  $L$  at different  $D/D_0$  values. Boundary conditions as in Fig. 2.



**Fig. 6.** Predicted methane flux  $F$  (cf. Eq. (2)) distribution within soil layer at different  $D/D_0$  values. Boundary conditions as in Fig. 2.



**Fig. 7.** Predicted distribution of the ratio of methane consumption  $Q$  (in the layer  $x$  to  $L$ ) to methane flux at the soil layer bottom ( $F_{x=L}$ ) at different  $D/D_0$  values. (=Per cent of oxidation of the methane stream). Boundary conditions as in Fig. 2.

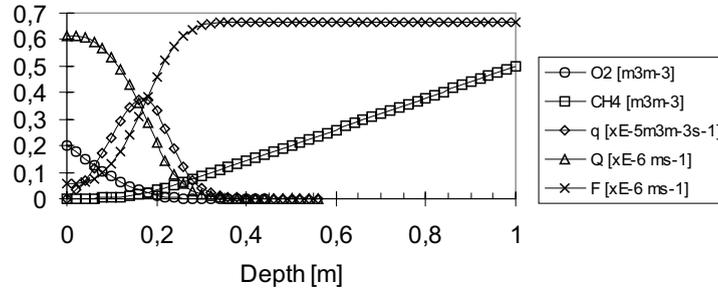
0.02, respectively. All the curves show a similar pattern in both figures but the absolute values of  $F$ ,  $Q$  and actual methanotrophic activity differ considerably, with only little differentiation in the concentration distribution of both substrates of the methane oxidation reaction.

Both the methane flux density at the bottom  $F_{x=L}$  and at the surface  $F_{x=0}$  of the soil layer (Fig. 10) increase with gas diffusivity. The depth at which methanotrophic activity reaches its maximum is only to a minor degree related to gas diffusivity and the variability is within several centimeters (16-19 cm) for the entire  $D/D_0$  range.

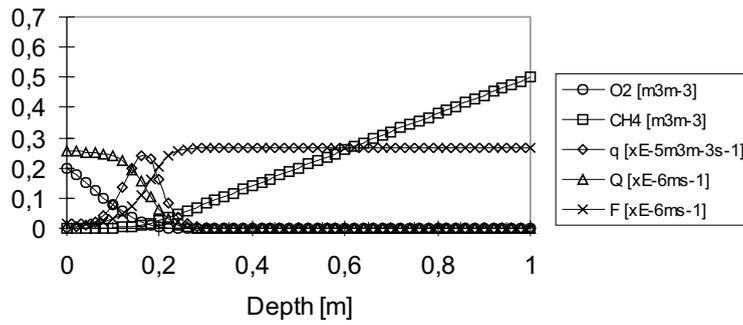
The ratio of the total methane consumption in the entire soil layer  $Q_t$  to the original methane

flux entering the soil  $F_{x=L}$  (Fig. 11) decreases from 1 (100% in the figure) at low  $D/D_0$  values to 0.7 at  $D/D_0=0.2$ . On the other hand the ratio of the highest effective methanotrophic activity (maximum value in Fig. 4) to the  $q_{max}$  value increases from about 0.1 at the lowest diffusivity levels to about 0.6 at  $D/D_0 = 0.2$ . It should be stated, that the latter ratio reflects the efficiency of utilization of the potential methanotrophic potential of the soil layer. Thus, it can be stated that this potential is actually utilized only in the range 10-60% in the most active part of the methanotrophic zone, while below and above it, the real activity is always lower.

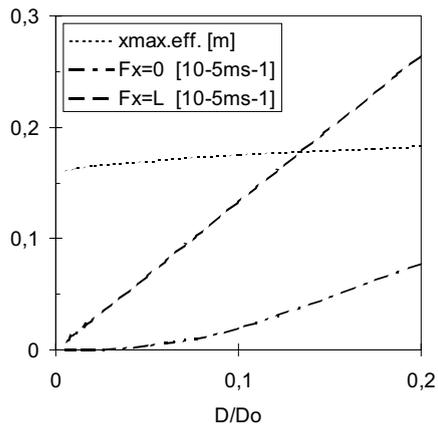
Total consumption of methane in the soil layer (Fig. 12), increases with  $D/D_0$  to almost



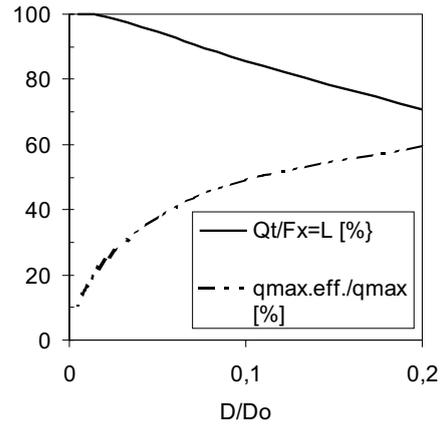
**Fig. 8.** Predicted distribution of methane and oxygen concentration, methanotrophic activity  $q$ , methane consumption  $Q$ , and the methane flux  $F$  at  $D/D_0$  value = 0.05. Boundary conditions as in Fig. 2. Vertical axis as in the legend.



**Fig. 9.** Predicted distribution of methane and oxygen concentration, methanotrophic activity  $q$ , methane consumption  $Q$ , and the methane flux  $F$  at  $D/D_0$  value = 0.02. Boundary conditions as in Fig. 2. Vertical axis as in the legend.



**Fig. 10.** Predicted depth of the highest methanotrophic activity ( $x_{max.eff}$ ) and the methane flux at the bottom of the soil layer ( $F_{x=L}$ ) and on its surface ( $F_{x=0}$ ) as a function of  $D/D_0$ . Boundary conditions as in Fig. 2. Vertical axis as in the legend.



**Fig. 11.** Predicted ratio of the total methane consumption in the soil layer  $Q_t$  to the initial methane stream at the soil bottom  $F_{x=L}$  and the ratio of the highest effective methanotrophic activity ( $q_{max.eff}/q_{max}$ ) as a function of  $D/D_0$ . Boundary conditions as in Fig. 2. Vertical axis as in the legend.

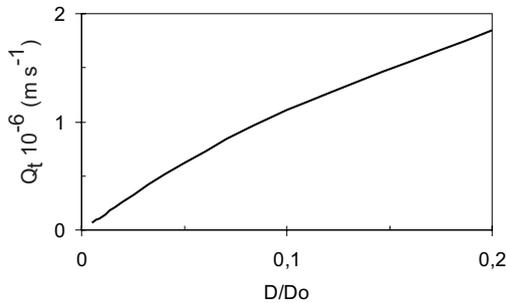


Fig. 12. Predicted total methane consumption in the soil layer ( $Q_t$ ) as a function of  $D/D_0$ . Boundary conditions as in Fig. 2.

$2 \cdot 10^{-5} \text{ m}^3 \text{ m}^{-2} \text{ s}^{-1}$ . This corresponds to the oxidation of  $0.173 \text{ m}^3 \text{ m}^{-2} \text{ d}^{-1}$  (or  $1730 \text{ m}^3 \text{ ha}^{-1} \text{ d}^{-1}$ ) which is comparable with the maximum value ( $0.22 \text{ m}^3 \text{ m}^{-2} \text{ d}^{-1}$ ) found experimentally by Kightly *et al.* [4].

#### CONCLUSIONS

1. Methane concentration in the soil is not highly dependent on  $D/D_0$ ; the changes within 4% range are possible only at 10-20 cm depth in the entire  $D/D_0$  range.

2. Soil oxygenation depth at 50% of the highest value of  $q_{max}$  in literature varies from 20 cm (at  $D/D_0=0.005$ ) to 50 cm (at  $D/D_0=0.2$ ).

3. Methanotrophic activity  $q$  (at a given constant value of  $q_{max}$ ) is concentrated within the oxic part of the soil, it increases with  $D/D_0$

and reaches the maximum value at the depth 16-20 cm; the maximum value increases with  $D/D_0$  and approaches 60% of  $q_{max}$  at  $D/D_0 = 0.2$ .

4. Cumulative methanotrophic capacity of the soil layer increases with the gas diffusivity up to almost  $2 \cdot 10^{-5} \text{ m}^3 \text{ m}^{-2} \text{ s}^{-1}$  at  $D/D_0 = 0.2$ .

5. Gas diffusion coefficient is a very important soil physical parameter decisive for the methanotrophic capacity of the soil layer covering landfill.

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