CALCULATION OF LANDFILL EMISSIONS CONSIDERING METHANE OXIDATION

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Abstract. *Sanitary landfills emit methane into the atmosphere, a gas that has a global warming potential 23 times higher than that of carbon dioxide. Reducing methane emissions is important from an environmental standpoint. In this work we developed an one-dimensional transport model for estimating methane emissions in landfills with explicit accounting for methane oxidation through pre-calculated oxidation coefficients. The methane concentration is obtained for the landfill and top soil cover regions. The methane oxidation coefficients were estimated based on experimetal data found in the literature. They lie between* $7x10^{-7}$ s^{-1} *and* $2x10^{-4}$ s^{-1} *for the top soil cover material and the suggested average value is* $3x10^{-6}$ s^{-1} *for a 30 cm depth inside the top soil cover. The suggested value for the municipal solid waste region is* $1.1x10^{-6}$ s^{-1}*. Sensititivty studies were performed and showed that the emission rates are strongly dependent on the oxidation coefficient utilized. The methane emission obtained for the CTR-Caieiras landfill, away from extraction wells, is* $2x10^{-5}$ *mol* CH_4/m^2s *for a 4 year old deposited wastes.*

Keywords*: landfill, methane, biogas, emission*

1. INTRODUCTION

Landfill emissions of biogas are important sources of greenhouse gases that contribute to global warming. About 50 % of the biogas volume produced in landfills is methane, which has a global warming potential 23 times greater than CO_2. Landfill emissions are usually measured at the landfill surface or estimated through calculations (Stein et al, 2001; Perera et al, 2002a; De Visscher and Van Cleemput, 2003, Hettiarachchi et al, 2007). For accurate calculations it is necessary a mathematical model that accounts for the methane production, its transport across the different landfill regions toward the atmosphere interface, and its consumption through oxidation (Stein et al. 2001; Perera et al. 2002a; De Visscher and Van Cleemput, 2003 ; Durmusoglu et al. 2005; Im et al., 2009, Yuan et al., 2009).

The emission from landfills is strongly affected by the oxidation rates in the cover soils due to the greater concentration of oxygen near the atmosphere interface (De Vischer et al., 1999; Stein et al., 2001; Miller and Clesceri, 2003; Perera et al 2002b; De Vischer and Van Cleemput, 2003). Several studies have been undertaken to determine methane oxidation in landfills including laboratory experiments which measured oxidation rates in sample materials (De Visscher et al., 1999; Stein et al., 2001; Perera et al 2002b; De Visscher and Van Cleemput, 2003, Gebert et al., 2003; Scheutz and Kjeldsen, 2004; Pawlowski and Stepniewsk, 2006; Jugni et al. 2008; Scheutz et al. 2008, Scheutz, et al. 2009; Abouchou et al., 2009; Im et al., 2009). An important fraction of the methane generated in a landfill is oxidized before reaching the atmosphere due to the high concentration of O_2 near the interface landfill-atmosphere.

The transport models need to account for the advection and diffusion or dispersion of the different gases present in the landfill, and the methane oxidation rate. Usually the methane oxidation rate is treated through a Michaelis-Menten equation that requires knowledge of both methane and oxygen concentrations. The Michalelis-Menten approach is a nonlinear relation that couples the two gas concentration equations in a manner that only numerical solutions are possible to be obtained even for simple homogenous problems (Stein et al, 2001; Perera et al, 2002a; De Visscher and Van Cleemput, 2003, Hettiarachchi et al, 2007).

This work aims at estimating the methane emission to the atmosphere through calculation taking into consideration the oxidation that occurs in the top part of the landfill. In order to facilitate the solution of the methane transport equation, we introduce a novel explicit coefficient to account for the methane oxidation that decouples the oxygen and methane equations. The approach is used to estimate the emissions from the CTR-Caieiras landfill in Sao Paulo, Brazil, and the results are compared with those of other landfills.

2. METHODS FOR ESTIMATING LANDFILL EMISSIONS

The important processes that occur in a landfill regarding the concentration of methane are its generation by biodegradation of organic materials, the transport of methane through the porous medium, and its sorption by oxidation reactions mediated by microorganisms. The one-dimensional steady-state balance equation in the vertical direction for the methane concentration is

$$-\frac{d}{dz}\left(D\frac{dC(z)}{dz}\right) + \quad A(z) = R(z). \tag{1}$$

where C(z,t) is the concentration of methane (mol CH_4/m^3), R(z,t) and A(z,t) are, respectively, the methane generation rate and consumption rate through oxidation (mol CH_4/m^3s), and D is the dispersion coefficient which represents the transport of methane molecules due to diffusion or dispersion process. Usually, the biogas in a landfill undergoes two different transport processes: advection and dispersion (Tchobanoglous et al. 1993). The advection process provides the flux of methane molecules due to pressure differences such as those caused by forced collection from landfill wells. The dispersion process, given by the Fick's law, provides the flux due to the scattering of methane molecules in the porous medium voids. In a landfill without forced gas collection the advection process is negligible and the transport process is controlled by the methane dispersion. Conversely, in a landfill with forced gas collection, the biogas transport is mainly controlled by the advective process.

Near vertical wells the emissions through the interface landfill-atmosphere are low because advection is the predominant mechanism for the biogas transport. The methane molecules move toward the vertical wells and are not emitted to the atmosphere. Away from the wells, we can consider only the one-dimensional vertical transport toward the atmosphere since the transverse velocity field is negligible. Thus the dispersion mechanism becomes the principal process of biogas transport and the advection term can be neglected, as is done in Eq. 1. The methane flux is then

$$J(z) = -D\frac{dC(z)}{dz}. \tag{2}$$

The methane oxidation rate is usually estimated using the Michaelis-Menten kinetics, which is a nonlinear relation involving the CH_4 and O_2 concentrations (Stein et al, 2001; Perera et al, 2002a; De Visscher and Van Cleemput, 2003, Hettiarachchi et al, 2007). In order to facilitate the solution of Eq. 1, the oxidation rate term is approximated by

$$A(z) = \sigma(z)\, C(z), \tag{3}$$

where σ(z) is an oxidation coefficient which depends on both methane and oxygen concentrations. In Section 5 we discuss how the oxidation coefficient is obtained. Substituting Eq. 2 in Eq. 1 we obtain

$$-\frac{d}{dz}\left(D\frac{dC(z)}{dz}\right) + \quad \sigma(z)C(z) = R(z). \tag{4}$$

To solve Eq. 4 is necessary to know the parameters that describe the landfill (D and σ), obtain estimates for the methane generation rate, R(z), and define boundary conditions. Eq. 4 has an explicit term to account for the oxidation rate of methane in the landfill. σ(z) may vary as a function of position but can be considered uniform within a region where the CH_4 and O_2 concentrations do not change very much. Thus, σ can be calculated in advance for several regions and corrected iteratively in case of significant differences between the obtained concentrations of O_2 and CH_4 and those used for estimating σ. Having an explicit oxidation coefficient in Eq. 4 allows easier solutions for the methane transport problem. This is the approach taken in this work.

2.1. The landfill one-dimensional model

The landfill is modeled according to Figure 1 with two different homogenous regions: the bottom region filled with the MSW material and the top region with the soil cover material. We consider two situations for estimating the landfill emissions: in the "A" configuration, we suppose that the landfill has no soil cover on the top, i.e., the MSW material is in direct contact with the atmosphere; in the "B" configuration, we suppose that there is a soil layer covering the landfill. At the bottom, the landfill has a sealing membrane that does not allow the methane to escape out. This model represents a landfill with a protecting layer at the bottom, which does not allow gases to escape or ingress, and on the top, with a porous surface that allows gases to ingress or escape. The boundary conditions considered are methane flux equals zero at the bottom and methane concentration equals zero at the interface with the atmosphere. In this case, we consider that sufficiently strong winds remove the methane molecules from that interface and keep its concentration zero there. For the "B" configuration we impose that the methane concentration and flux are continuous at z = H.

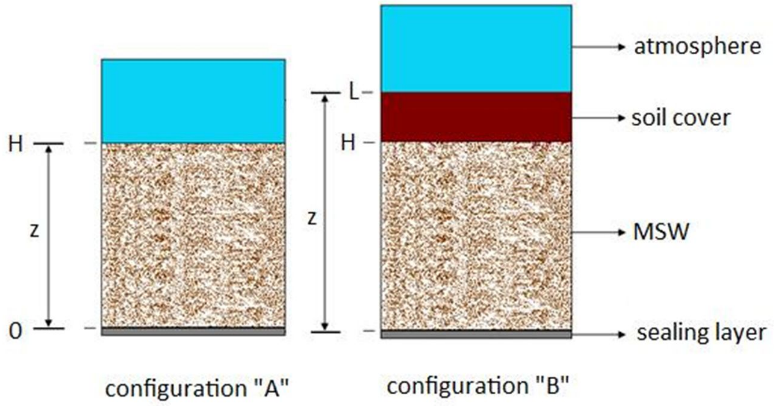

Figure 1. Vertical schematic for the CTR-Caieiras landfill

2.2. Methane generation rate and dispersion coefficients

The landfill considered in this analysis is the CTR-Caieiras located in the municipality of Caieiras (23°21'51" S and 46º44'26" W) in São Paulo state, Brazil and owned by Essencis Soluções Ambientais SA. The landfill is part of a waste treatment facility aiming at disposing industrial and municipal solid wastes from Sao Paulo and other surrounding cities. The landfill receives approximately 8,000 tMSW/day, operates in a daily basis and its general procedure is to receive the material from trucks, and deposit, spread and compact the waste material. At the end of each day, a 0.5 m layer of soil is deposited to cover the waste. The annual temperature lies between 16 and 22^{o}C.

The rate of methane generation in the landfill in the landfill is considered uniform across the whole region. The landfill first order methane generation parameters are L = 4150 mol/t MSW and k = 0,093 year^{-1} (Candiani, 2010). The MSW material specific mass, ρ_{MSW}, is 0.6 tMSW/m^3 and its porosity, ϕ, is 0.3. The volumetric methane generation rate is approximated by a first order model

$$R(z) = R_0 = \frac{kL\rho_{MSW}}{\phi_R} e^{-kt} \tag{5}$$

where t is the time elapsed since the MSW was deposited in the landfill (4 years). The methane generation rate, R_0, is $2.45\mathrm{x}10^{-5}$ mol CH_4/m^{3}s.

The dispersion coefficient of landfill materials has been studied by several researchers (Perera et al. 2002b; Van Cleemput and Visscher, 2003; Clesceri and Miller, 2003; Im et al. 2009; Abichou et al. 2009). The biogas dispersion in porous media such as the landfill material is slower than in the air due to its reduced free volume and the medium tortuosity. The dispersion coefficient in a porous media is obtained from (Perera et al. 2002b; Van Cleemput and Visscher, 2003; Clesceri and Miller, 2003; IM et al. 2009; Abichou et al. 2009; Silva, 2010)

$$D = \gamma D_{air} \tag{6}$$

where D_{air} is the methane diffusion coefficient in the air and the factor γ is given by

$$\gamma = \left(\frac{\phi - c}{1 - c}\right)^d \tag{7}$$

where ϕ is the porosity, and c and d are constants (Perera et al. 2002b; Van Cleemput and Visscher, 2003; Clesceri and Miller, 2003; Im et al. 2009; Abichou et al. 2009; Silva, 2010). The methane diffusion coefficient in the air and suggested values for the constants c and d are presented in Table 2. These parameters were used to estimate the methane dispersion coefficients for the MSW and soil cover materials utilizing Eqs. 6 and 7. The obtained reduction factor, γ, and the dispersion coefficients for the MSW and soil cover materials are also presented in Table 2. The subscripts R and C denote the MSW and soil cover regions.

3. OXIDATION COEFFICIENT ESTIMATION

Several studies have been undertaken to determine methane oxidation in landfills including laboratory experiments which measured oxidation rates in landfill sample materials (De Visscher et al., 1999; Stein et al., 2001; Perera et al 2002b; De Visscher and Van Cleemput, 2003, Gebert et al., 2003; Scheutz and Kjeldsen, 2004; Pawlowski and Stepniewsk, 2006; Jugnia et al. 2008; Scheutz et al. 2008, Scheutz, et al. 2009; Abouchou et al., 2009; Im et al., 2009). An important fraction of the generated methane is oxidized before reaching the atmosphere. Near the interface with the atmosphere, there is a high concentration of O_2 and the CH_4 oxidation by microorganisms may be substantial. Many different parameters affect the methane oxidation such as temperature, pH, humidity, concentration of O_2 and others gases (Stein et al. 2001; Scheutz and Kjeldsen, 2004; Jugnia et al. 2008; Scheutz et al. 2008, Scheutz, et al. 2009). Temperatures varying between 25 and 35 ^oC degrees are ideal for methane oxidation in landfills. Temperatures below 10 °C drastically reduce methane oxidation, and above 35 °C inhibit the microorganism's mediation for methane oxidation.

Several research groups present experimental results about oxidation rates in terms of the Michaelis-Menten kinetics (De Visscher et al., 1999; Stein et al., 2001; Perera et al 2002b; De Visscher and Van Cleemput, 2003, Gebert et al., 2003; Pawlowski and Stepniewsk, 2006; Abouchou et al., 2009; Im et al., 2009). In terms of this formalism σ(z,t) is defined as

$$\sigma(z) = \frac{V_m\, O(z)}{\big(K_C + C(z)\big)\big(\,K_O + O(z)\big)} \tag{8}$$

where V_m is the maximum methane oxidation rate, K_C and K_O are saturation constants for CH_4 and O_2, respectively, and O(z) and C(z) are the O_2 and CH_4 concentrations.

The soil cover material considered in these research works presented specific masses between 1270 kg/m^3 and 2650 kg/m^3. The maximum oxidation rates values, V_m, obtained by these authors ranged from 2.3x10^{-6} to 7.6x10^{-3} mol/m^{3}s. Gebert et al. (2003) in their experiments obtained V_m value of 4.94x10^{-4} mol/m^{3}s and observed that the methane oxidation was significant for O_2 concentrations above 0.76 mol/m^3. Abichou et al. (2009) suggest a V_m value of 2x10^{-3} mol/m^{3}s for compost cover layers, and 5x10^{-4} mol/m^{3}s for regular soil covers. The saturation constants K_C and K_O of Eq. 8 obtained by several researchers also present large variation. K_C values range from 0.01 to 2.01 mol/m^3, while K_O values range from 0.03 to 4.7 mol/m^3.

Scheutz et al. (2008) present results of methane oxidation rates and the concentration of methane and other gases as a function of depth determined in experiments performed in the Grand'Landes landfill in France. The measurements were performed in environmental conditions similar to those of the CTR-Caieiras landfill. The temperature in the cover measured 0.1 m below the surface ranged from 17 to 25 °C, the moisture content of the landfill ranged from 0.13 to 0.19, and the pH 5.5 to 7.6. Since the experiments presented the oxidation rate and the oxygen and methane concentrations as a function of depth, it was possible to obtain the oxidation coefficient utilizing Eq. 8.

Figure 2 presents the methane oxidation coefficient derived from the experimental results of these research groups for conditions which approximate three different depths in the soil cover layer. The 5, 30 and 55 cm depths correspond to pairs of O_2 and CH_4 concentrations of 8.25 and 3 mol/m^3, 4 and 10 mol/m^3 and 0.3 and 48 mol/m^3, respectively.

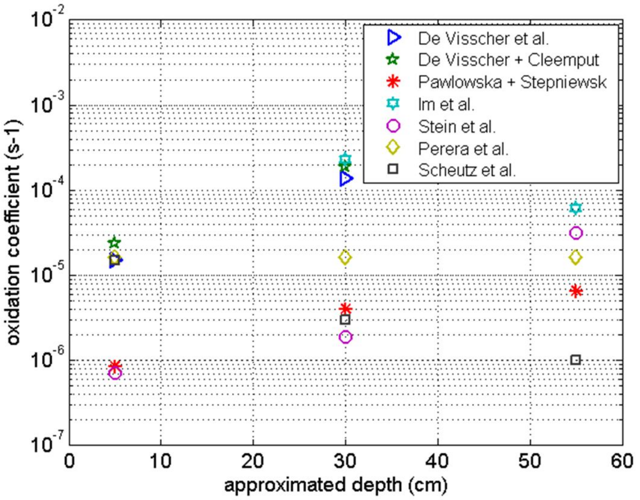

Figure 2. Oxidation coefficient, σ, based on the Michaelis-Menten formalism according to reported experimental data

The obtained oxidation coefficients, using Eq. 8, present large variation according to the experimental Michaelis-Menten data reported by the several authors. Near the atmosphere interface σ varies from 7×10^{-7} to 3×10^{-5} s^{-1}; at about 30 cm depth, the σ varies from 2×10^{-6} to 3×10^{-4} s^{-1}; and at about 55 cm depth, where the oxygen concentration is low, σ varies from 1×10^{-6} to 6×10^{-5} s^{-1}.

The oxidation coefficients adopted for the CTR-Caieiras soil cover material are those obtained from the Grand'Landes landfill in France (Scheutz et al., 2008) due to its close environmental conditions. The adopted value to represent the soil cover material is the one at 30 cm depth, 3×10^{-6} s^{-1}. The oxidation coefficient estimated for the MSW region is 1.1×10^{-6} s^{-1}, which corresponds to the lowest value for the 55 cm depth presented in Figure 2. This figure was also chosen because it produces an internal pressure in the bottom of the landfill around 1 atm, an expected value for an equilibrium landfill-atmosphere system. The choices can be considered arbitrary given the large spread of the data presented in Figure 2. Thus, some sensitivity study shall be done to verify the impact of such a choice on emission results. Table 2 shows the oxidation coefficients for the soil and MSW regions adopted for the CTR-Caieiras landfill.

Table 2. Parameters describing the CTR-Caieiras landfill

Parameter	Value
ϕ_R - MSW porosity	0.30
ϕ_C – soil cover porosity	0.22
Methane diffusion coefficient in the air (De Visscher et al., 1999)	$2.12 \times 10^{-5} m^2/s$
c parameter (Perera et al., 2002b)	0.15
d parameter (Perera et al., 2002b)	1.1
γ factor for the MSW material	0.113
γ factor for the soil cover material	0.075
D_R - dispersion coefficient in the MSW	3.14×10^{-6} m^2/s
D_C - dispersion coefficient in the soil cover	1.36×10^{-6} m^2/s
σ_R – oxidation coefficient for MSW region	1.1×10^{-6} s^{-1}
σ_C – oxidation coefficient for soil cover region	3×10^{-6} s^{-1}

4. METHANE EMISSION RESULTS AND ANALYSIS

The first calculation is for the "A" configuration which represents a landfill without any cover on the top and with the MSW refuse in direct contact with the atmosphere. Since the MSW region is considered homogenous, with uniform transport parameters D_R and σ_R, and the methane generation rate, $R(z) = R_0$, is also uniform, Eq. 4 can be analytically solved. For boundary conditions of the zero flux at the z = 0 and zero concentration at z = H, the solution is

$$C(z) = \frac{R_0}{\sigma_R}\left(1 - \frac{\cosh(\beta_R z)}{\cosh(\beta_R H)}\right) \tag{9}$$

where $\beta_R = \sqrt{\sigma_R / D_R}$. Eq. 9 gives the distribution of CH_4 concentration in the landfill. The methane concentration is maximum and uniform at the landfill bottom. At about 10 m depth, the methane concentration already reaches its maximum value. Near the interface with the atmosphere it approaches to zero representing the case in which a sufficiently strong wind removes the methane molecules from the landfill-atmosphere interface.

For the "B" configuration, we need to solve Eq. 4 for the bottom ant top regions. The boundary conditions are J(0) = 0, C(L) = 0, and the continuity of concentration and flux at the interface z = H. The generation term, R(z), equals zero at the top soil region. If the subscripts R and C indicate the MSW and soil cover regions, the solution is

$$C(z) = \begin{cases} \dfrac{R_0}{\sigma_R}\left(1 - A_R \cosh(\beta_R z)\right), & for\ \ 0 \le z \le H, \quad (MSW\ region) \\ \dfrac{R_0}{\sigma_R} A_C\left(e^{\beta_C (z-2L)} + e^{-\beta_C z}\right), & for\ \ H \le z \le L, \quad (soil\ cover\ region) \end{cases} \tag{10}$$

where A_R and A_C are constants (Silva, 2010), $\beta_R = \sqrt{\sigma_R / D_R}$, and $\beta_C = \sqrt{\sigma_C / D_C}$. Figure 3 shows the methane concentration for the "A" and "B" configurations. The soil cover causes the concentration to increase at the top part of the landfill.

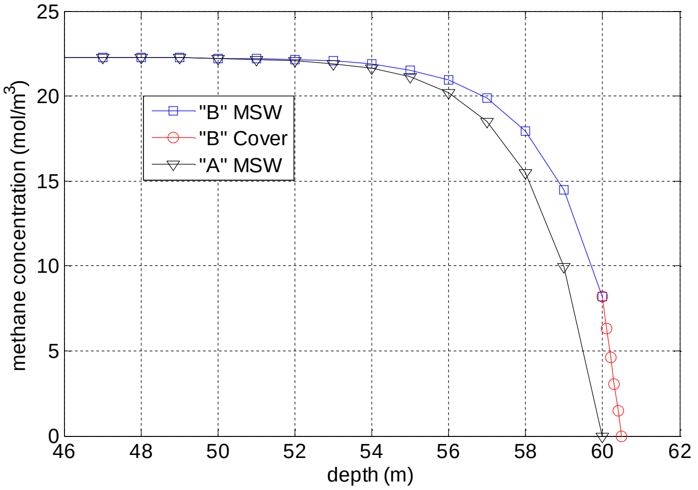

Figure 3. Methane concentration as function of depth for the “A” and “B” configurations

Figure 4 shows the methane flux as a function of landfill depth obtained using Eq. 2. The methane flux is zero at the base of the landfill, increases as it approaches the interface with the atmosphere, and reaches 4.2x10^{-5} mol/m^{2}s for the “A” configuration and 2x10^{-5} mol/m^{2}s for the “B” configuration. This means that the CH$_4$ emission rates to the atmosphere are 4.2x10^{-4} and 2x10^{-5} mol/m^{2}s for the “A” and “B” configurations, respectively. The methane flux toward the atmosphere for the configuration without cover is twofold that with a 0.5 m top soil cover.

Although the methane concentration in the landfill-atmosphere interface was set to zero as a boundary condition, there is a large flux of molecules moving from the landfill to the atmosphere. Physically, the methane transferred to the atmosphere is readily removed by winds on the surface so that its concentration is always negligible.

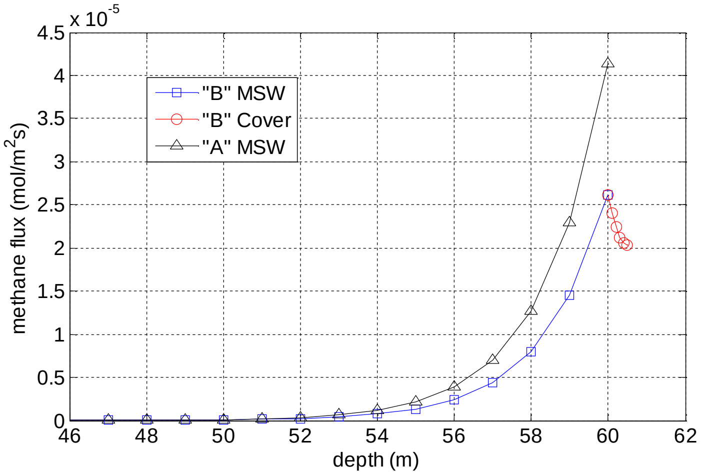

Figure 4. Methane flux as function of depth for the “A” and “B” configurations

In order to assess the methane emission sensitivity to the top soil oxidation coefficient, we performed calculations with different values: 7x10^{-7} s^{-1}, 3x10^{-6} s^{-1} and 4.5x10^{-5} s^{-1}. Figure 5 shows the methane flux as a function of depth for two extreme values of oxidation coefficient. Larger σ increases the flux at the top of the MSW region but decreases it in the top soil region due to the stronger oxidation rate. The results indicate that σ larger than 10^{-4} s^{-1} would decrease the methane flux to the atmosphere to negligible values. It seems that top soils should have oxidation coefficients between 10^{-6} and 10^{-4} s^{-1}. The oxidation coefficient plays an important role in methane emissions to the atmosphere.

Table 3 presents field measurements in different landfills and the calculated results obtained in this work. Emission rates vary a lot in large landfills and many authors present minimum, medium and maximum values in their field measurements. Since our calculations do not consider forced methane collection, the results represent maximum emissions. The calculated values have the same order of magnitude of measured values. Maciel (2003) and Mariano

(2008) data represent examples of almost uncovered landfills but their results are 6 times greater than the “A” configuration model which does not have any top soil cover. The other data, with thicker top soil cover, seem closer to the calculated result with oxidation coefficient equals to $3\text{x}10^{-6}$ s^{-1}, in spite of different top soil thickness. cover (0.5 m). The calculated results show that the methane fluxes can be approximated with such oxidation coefficient, but more meaningful comparisons need to consider measurement conditions, soil cover, and solid waste age and characteristics.

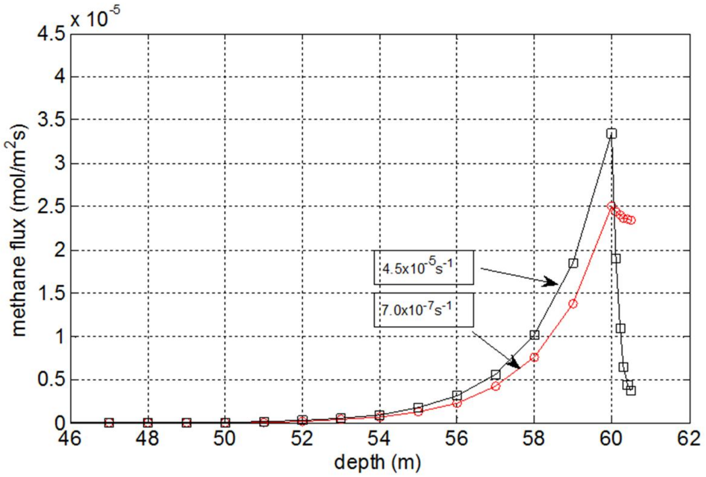

Figure 5. Methane flux as a function of depth for different values of oxidation coefficient, σ

Table 3 - Values of methane flux measured at various landfills and calculated by the model developed here

	Soil cover thickness (m)	Methane flux (mol $CH_4/m^2.s$)
Maciel (2003)	0.2 – 0.7	2.8×10^{-4}
Mariano (2008)	0.2 – 0.9	2.6×10^{-4}
Christophersen et al. (2001)	1	2.4×10^{-5}
Scheutz et al. (2008)	1	2.1×10^{-5}
Abichou et al. (2009)	0.8	4.2×10^{-5}
“A” configuration (calculated)	0	$4.2\text{x}10^{-5}$
$\sigma = 7\text{x}10^{-7}$ s^{-1} (calculated)	0.5	$2.3\text{x}10^{-5}$
$\sigma = 3\text{x}10^{-6}$ s^{-1} (calculated)	0.5	$2.0\text{x}10^{-5}$
$\sigma = 4.5\text{x}10^{-5}$ s^{-1} (calculated)	0.5	$3.5\text{x}10^{-6}$

5. CONCLUSION

In this work we developed an one-dimensional transport model for estimating the methane emission rates in landfills. In the model the oxidation rate is estimated through an oxidation coefficient obtained from experimental data available in the literature. We obtained solutions for the methane concentration inside the landfill for two different configurations: with and without a top soil cover.

The methane oxidation coefficient, σ, for different top soil cover depths lies between $7\text{x}10^{-7}$ s^{-1} and $2\text{x}10^{-4}$ s^{-1}. For the MSW was considered an oxidation coefficient of $1.1\text{x}10^{-6}$ s^{-1}. A sensitivity analysis was performed for different values of methane oxidation coefficients for the top soil cover material. The results showed that for the range cited above the methane flux toward the atmosphere can vary from negligible to $3\text{x}10^{-5}$ mol CH_4/m^2s. The suggested average oxidation coefficient for usual top soil materials is $3\text{x}10^{-6}$ s^{-1}, corresponding to a 30 cm depth. The model was used to estimate the methane emission rate for the CTR-Caieiras landfill. The estimated emission rate was a methane flux of $2\text{x}10^{-5}$ mol CH_4/m^2s for the sites with MSW age of about 4 years.

These results were compared to measurements presented in the literature for different landfills. For almost uncovered landfills, the results were an order of magnitude smaller; for covered landfills the resulted agreed well. As a future work, we intend to perform detailed comparisons with experimental emission rates.

6. ACKNOWLEDGEMENTS

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