

Approximate Estimation of Landfill Emissions Considering Methane Oxidation

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Abstract: An approximate analytical scheme is presented to estimate landfill methane emissions taking into account the oxidation that occurs in the soil cover. To facilitate the solution of the methane transport equation we introduce a region dependent coefficient to account for the methane oxidation. Expressions for the distribution of methane concentration and methane flux inside the landfill and cover regions are obtained. The approach was applied to the CTVA-Caieiras landfill which was modeled as a vertical one-dimensional landfill with homogenous solid waste and soil cover regions. The methane emission obtained for the landfill was $2 \times 10^{-5} \text{ mol m}^{-2} \text{ s}^{-1}$ for a 4 year old deposited waste with a 0.5 m soil cover. The calculation compared well with the median of 11 field measurements conducted at the CTVA-Caieiras. The emission rates were strongly dependent on the oxidation coefficient utilized, which varies with the cover material and microclimate conditions of the site. The oxidation coefficient can be interpreted as the probability per unit of time of methane oxidation in the medium. The scheme provides a qualitative description of the methane transport and oxidation phenomena in landfills.

Keywords: Biogas, emission, Landfill cover, methane, oxidation, oxidation coefficient.

INTRODUCTION

Landfill emissions of methane are important sources of greenhouse gases that contribute to global warming. Methane generated through biodegradation of municipal solid waste (MSW) migrates through the landfill and its soil cover before reaching the atmosphere. Near the interface with the atmosphere the presence of high concentrations of oxygen allows for methane oxidation mediated by microorganisms [1-4]. These emissions are usually measured at the landfill surfaces or estimated through calculations based on first order models for methane generation, methane transport across the different landfill regions toward the atmosphere, and methane oxidation [1,5].

The general approach adopted for calculation of methane emissions from landfills is to consider average transport parameters for different landfill regions, and a more detailed treatment for the oxidation rate in the cover region. This important methane sorption process depends on different variables such as microclimate conditions, temperature, atmospheric pressure, moisture, soil conditions, concentration of O_2 and CH_4 , and population of methanotrophs and their activities [2,3,4]. Calculation models usually treat the methane oxidation rate through a Michaelis-Menten equation that requires knowledge of both

methane and oxygen concentrations inside the landfill, and specific microclimate and cover soil conditions. Therefore the results are valid for those conditions and may change during the year or even during night and day [1-5].

The Michaelis-Menten equation couples the gas concentration equations for oxygen and methane so that only numerical solutions are possible for the methane transport problem in landfills, even for simple homogenous problems [1,2,5,6]. This article aims at obtaining an approximate analytical solution for the problem of methane emission from landfills to the atmosphere taking into consideration the oxidation that occurs in their soil covers. To facilitate the solution of the methane transport equation we introduce a homogenous oxidation coefficient for the cover region that decouples the oxygen and methane equations. This approximation allows obtaining analytical expressions for the methane concentration inside the landfill and the cover regions, and the methane flux to the atmosphere. The approach, considering a one-dimensional landfill model, is used to estimate the emissions from the CTVA-Caieiras landfill in São Paulo, Brazil [7]. We start introducing the methane transport equation with an explicit oxidation coefficient, present the scheme adopted to obtain it from the literature data and the model describing the CTVA-Caieiras, and then present the results, discussions, and conclusions.

METHODS AND DATA

To obtain the methane flux emitted from a landfill to the atmosphere is necessary to solve the transport equation for

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the methane concentration in the vertical direction (landfill depth). We modeled the landfill in the vertical direction dividing it in two regions: the MSW region and the top soil cover region which interfaces with the atmosphere. This two-region system is solved to furnish the methane flux to the atmosphere. To obtain an analytical solution we introduce an explicit methane sorption coefficient to account for the oxidation in the cover region. The approach taken in this work can be summarized as follows: a) introduce a homogenous oxidation coefficient to decouple the equations for the oxygen and methane concentrations; b) obtain estimates for the oxidation coefficient from the literature data based on the Michaelis-Menten kinetics parameters; c) solve the transport equation for the methane concentration for a two-region landfill model (MSW region and soil cover region); and d) compare the results with field measurements of methane flux conducted at the CTVA-Caieiras landfill.

METHANE TRANSPORT EQUATIONS WITH AN EXPLICIT OXIDATION COEFFICIENT

The one-dimensional steady-state balance equation for the methane concentration in the vertical direction can be written as

$$\frac{dJ(z)}{dz} + A(z) = R(z) \quad (1)$$

where $J(z)$ is the methane flux ($\text{mol m}^{-2}\text{s}^{-1}$), $R(z)$ is the methane generation rate ($\text{mol m}^{-3}\text{s}^{-1}$), and $A(z)$ is methane sorption rate through oxidation and other means ($\text{mol m}^{-3}\text{s}^{-1}$) [7,8]. For problems in which the advection term is negligible the methane flux can be described by the Fick's law

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

where $C(z)$ is the methane concentration (mol m^{-3}), and D is the dispersion coefficient ($\text{m}^2 \text{s}^{-1}$) [7,8]. The methane sorption rate through oxidation, $A_{ox}(z)$, is usually estimated using the Michaelis-Menten kinetics, which is a nonlinear relation involving the CH_4 and O_2 concentrations [1,2,3,5,6], i.e.,

$$A_{ox}(z) = \frac{V_m O(z) C(z)}{(K_C + C(z))(K_O + O(z))} \quad (3)$$

where V_m is the maximum methane oxidation rate bearing information about the microclimate, soil properties, methanotroph population, and other environmental conditions, K_C and K_O are the half saturation constants for CH_4 and O_2 , respectively, and $O(z)$ and $C(z)$ are the distributions of O_2 and CH_4 concentrations.

In sites with gas extraction it is necessary to account for transversal gas migration toward the extraction wells which lower the CH_4 concentration in the bottom of the landfill region. This reduces the CH_4 concentration gradient in the vertical direction, and consequently reduces the diffusive methane flux to the atmosphere [4,5,9]. In this approach the transversal gas migration is accounted for with a fictitious transversal sorption rate term, $A_T(z)$, so that the total methane sorption rate is given by

$$A(z) = A_T(z) + A_{ox}(z). \quad (4)$$

To decouple the gas concentration equations we introduce an explicit oxidation coefficient in the definitions of $A_{ox}(z)$ and $A_T(z)$, i.e.,

$$A(z) = \sigma(z) C(z) \text{ with } \sigma(z) = \sigma_T(z) + \sigma_{ox}(z) \quad (5)$$

where $\sigma(z)$ is the total sorption coefficient, $\sigma_T(z)$ is the transversal sorption coefficient and $\sigma_{ox}(z)$ is the oxidation coefficient. Using Eqs. 3, 4 and 5 we obtain an expression for the oxidation coefficient,

$$\sigma_{ox}(z) = \frac{V_m O(z)}{(K_C + C(z))(K_O + O(z))}. \quad (6)$$

Substituting Eqs. (2) and (5) into Eq. (1) we obtain for the methane concentration equation

$$-\frac{d}{dz} \left(D \frac{dC(z)}{dz} \right) + \sigma(z) C(z) = R(z). \quad (7)$$

To obtain a solution for the methane concentration is necessary to simultaneously solve Eq. 7, a similar equation for the oxygen concentration, and Eq. 6 which couples the two previous equations. While this is usually carried out numerically, Eq. 7 can be analytically solved for the methane concentration if the diffusion coefficient, D , and the sorption coefficient, σ , are considered homogenous in a given region. Imposing homogenous transport parameters reduces the practical application of the results but allows physical insights to the problem of methane oxidation in soil covers which are the aim of this article. The sorption coefficients σ_{ox} and σ_T have units of inverse time (s^{-1}) and can be interpreted, respectively, as probabilities per unit of time for methane oxidation and methane escape from the landfill through the collection wells.

METHANE FLUX MEASUREMENTS

The accuracy of the scheme presented in the previous section is verified against field measurements conducted at the CTVA-Caieiras landfill located in the municipality of Caieiras (23°21'51" S and 46°44'26" W) in São Paulo state, Brazil. The annual temperature in the region lies between 16 and 22°C. The landfill is part of a waste treatment facility aiming at disposing industrial and municipal solid wastes from São Paulo and other surrounding cities. The landfill receives approximately 7,000 t/day of MSW, and its general procedure is to collect the material from trucks, and then to deposit, spread and compact the waste at the site. At the end of each day a 0.5 m soil layer is used to cover the waste. The height of waste material can reach up to 60 m [7].

Table 1 presents the MSW material composition, and Table A1 in the Appendix presents the MSW porosity, and its first order kinetics parameters which allow estimating the methane generation rate at the site [7]. The porosity of the soil cover material is also presented in Table A1. The average waste moisture content is 60.9 %. The organic and moisture contents of the waste are high and similar to typical tropical regions, but the site has a highland climate controlled by the local relief. The average annual rainfall varies between 1250 and 1350 mm. Due to high elevation the mean annual temperature varies between 16 °C and 18 °C. The mean temperature during the summer varies between 19 °C and 22 °C and during the winter, between 13.5 °C and 15 °C [7]. In 2010 the landfill covered an area of 330,000 m² with 170 methane extraction wells. The site was

divided in three areas of waste deposition designated as Phases 1, 2 and 3.

Table 1. Composition of the CTVA-Caieiras MSW [7].

Material	Weight Fraction (%)
Organic	58.3
Plastic	15.2
Paper and cardboard	14.6
Glass	2.5
Ferrous metals	1.8
Textile, leather and wood	3.7
Others (soil and rubble)	3.9
Total	100

Methane flux measurements were conducted at Phase 2 which covered an area of 165,000 m². The measurement locations were chosen to be away from the 85 wells available in Phase 2. The distance between the wells is about 45 m. Eleven methane flux measurements were conducted using the flux chamber technique [4,5,7]. The chambers were constructed of galvanized plates with a size of 0.84x 0.79x 0.05 m (covering an area of 0.66 m² and volume of 33.2 L). During the measurements, the chamber was sealed to the ground by covering its border with earth material. Four methane samples were collected sequentially over a 4 min period using disposable syringes fitted with plastic stopcocks. Samples were analyzed on a gas chromatograph equipped with a flame ionization detector with a resolution of 10 ppb of CH₄. The methane flux was determined from concentration data plotted versus elapsed time [7].

THE VERTICAL LANDFILL MODEL

The CTVA-Caieiras landfill is modeled in one-dimensional geometry with two different homogenous regions: the bottom region filled with municipal solid waste material and depth $H = 60$ m, and the top region with the soil cover material with thickness of 0.5 m. Fig. (1) displays the two configurations considered in this article: in configuration A we assume the landfill has no soil cover on the top and the MSW material is in direct contact with the atmosphere; in configuration B we assume 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. At the top there is a porous surface that allows gases to ingress or escape [5,9].

The boundary condition at the landfill bottom is methane flux equals zero. At the interface with the atmosphere we adopt the boundary condition suggested by De Visscher and Van Cleemput [2]. At these interfaces the methane concentration can be 10 to 20 % higher than the atmospheric values (around $8 \times 10^{-5} \text{ mol m}^{-3}$) [10], but still very small compared with typical methane concentrations inside the landfill (around 15 mol m^{-3}). Thus the boundary condition was simplified and taken as negligible methane concentration. For configuration B we imposed that the

methane concentration and flux be continuous at the interface between the MSW and soil cover regions ($z = H$).

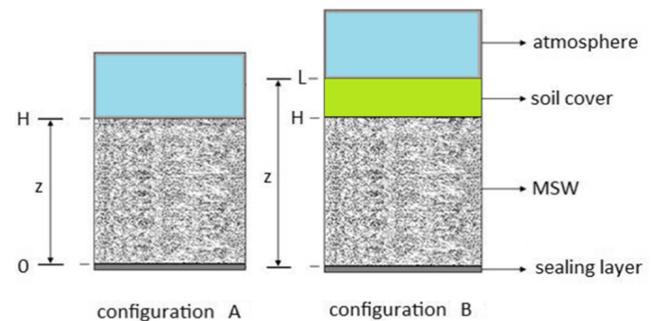


Fig. (1). Vertical schematic for the CTVA-Caieiras landfill.

Estimates for Transport Parameters

The methane generation rate was determined using first order kinetics parameters measured for the CTVA-Caieiras landfill [7]. The methane dispersion coefficients were determined using the procedure suggested by De Visscher and Cleemput [2], Perera *et al.* [11], Im *et al.* [12], Abichou *et al.* [13] and Silva [14] based on diffusion coefficients in the air and data about media porosity and tortuosity. The obtained dispersion coefficients are presented in Table A2 in the Appendix.

The literature presents methane oxidation rate data in landfill covers obtained in field and laboratory measurements. The data, usually formatted as parameters of the Michaelis-Menten kinetics equation, present very different rates of methane oxidation under different site and microclimate conditions [1, 2, 11-13, 15-19]. The soil cover material considered in these studies present specific masses between 1270 kg m^{-3} and 2.650 kg m^{-3} . The maximum oxidation rates values, V_m , of Eq. (3) obtained by these authors ranged from 2.3×10^{-6} to $7.6 \times 10^{-3} \text{ mol m}^{-3} \text{ s}^{-1}$. Gebert *et al.* [15] in their experiments obtained V_m value of $4.94 \times 10^{-4} \text{ mol m}^{-3} \text{ s}^{-1}$ and observed that the methane oxidation was significant for O_2 concentrations above 0.76 mol m^{-3} . Abichou *et al.* [13] suggest a V_m value of $2 \times 10^{-3} \text{ mol m}^{-3} \text{ s}^{-1}$ for compost cover layers, and $5 \times 10^{-4} \text{ mol m}^{-3} \text{ s}^{-1}$ for regular soil covers. The saturation constants K_C and K_O of Eq. 6 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.* [17] present results for methane oxidation rates and for concentrations of methane and other gases as a function of depth determined in experiments performed in the Grand'Landes landfill in France. The temperature in the cover region, measured 0.1 m below the surface, ranged from 17 to 25°C, the moisture content of the landfill ranged from 13 % to 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. 6. We considered three depths (5, 30 and 55 cm), and for each depth pairs of O_2 and CH_4 concentrations furnished by Scheutz *et al.* [17]; respectively 8.25 and 3 mol m^{-3} for 5 cm; 4 and 10 mol m^{-3} for 30 cm; and 0.3 and 48 mol m^{-3} for 55 cm.

Fig. (2) displays the methane oxidation coefficient obtained through Eq. 6 utilizing for K_C , K_O and V_m data from different literature, and pairs of oxygen and methane concentrations for these three different depths obtained from Scheutz *et al.* [17]. The obtained oxidation coefficients, using Eq. 6, 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 $3 \times 10^{-5} \text{ s}^{-1}$; at about 30 cm depth, the σ varies from 2×10^{-6} to $3 \times 10^{-4} \text{ s}^{-1}$; and at about 55 cm depth, where the oxygen concentration is low, σ varies from 1×10^{-6} to $6 \times 10^{-5} \text{ s}^{-1}$.

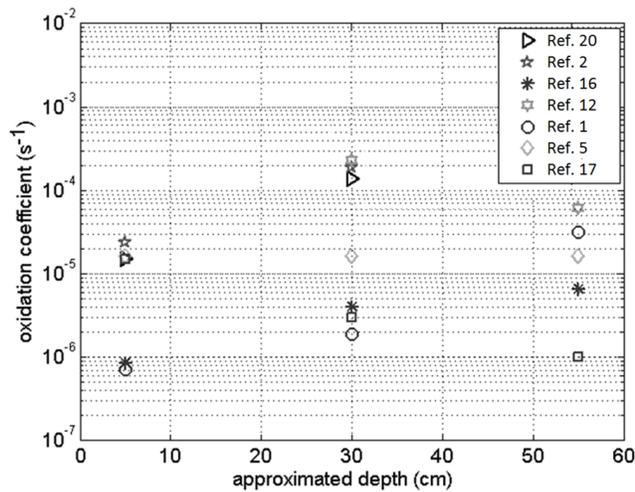


Fig. (2). Oxidation coefficient, σ , based on the Michaelis-Menten data reported in different literature. The variation is due to different site and microclimate conditions.

The oxidation coefficients adopted for the CTVA-Caieiras soil cover material are those obtained from the GrandLandes landfill in France [17] due to closer environmental conditions. The adopted value to represent the soil cover material is the one at 30 cm depth [14, 17]. The oxidation coefficient was considered negligible for the MSW region due to the lack of O_2 . All data required to obtain the transport parameters are presented in Table A2 in the Appendix.

The transversal migration of CH_4 and O_2 toward the extraction wells reduces the internal pressure inside the landfill which is expected to be around 1 atm for a landfill-atmosphere system close to equilibrium. The transversal sorption coefficient in the MSW region was taken as $1.1 \times 10^{-6} \text{ s}^{-1}$ in order to obtain this internal pressure in the bottom of the landfill [14]. In the cover region the transversal migration is expected to be less important because the extraction through the wells starts to occur about one meter below it. Thus in the cover region this transversal migration was neglected.

RESULTS

Calculated Methane Concentration Inside the Landfill

The analytical solution for the methane concentration of configurations A and B are presented in Table A3 in the Appendix. Since the MSW and cover regions are considered homogenous, with uniform dispersion coefficients, oxidation coefficients, and methane generation rate ($R(z) = R_0$), Eq. 7 could be analytically solved for both configurations. Fig. (3)

displays the methane concentration for configurations A and B. The 0.5 m soil cover causes the concentration to increase at the top part of the landfill. The methane concentration in the cover region varies from 8 mol m^{-3} to negligible values. For the average value of 4 mol m^{-3} we obtain an oxidation rate of $1.2 \times 10^{-5} \text{ mol m}^{-3} \text{ s}^{-1}$.

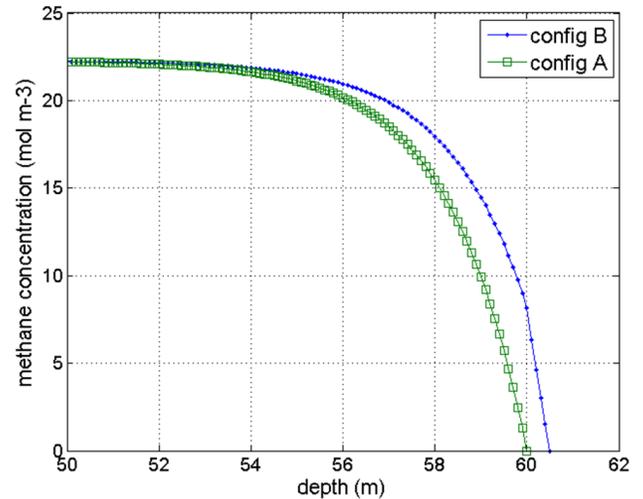


Fig. (3). Methane concentration as function of depth for configurations A and B.

Calculated Methane Flux to the Atmosphere

Fig. (4) shows the methane flux as a function of landfill depth for configurations A and B. The corresponding analytical expression for configuration B is present in Table A3 in the Appendix. The methane flux is zero at the base of the landfill, increases as it approaches the interface with the atmosphere, and reaches $4.2 \times 10^{-5} \text{ mol m}^{-2} \text{ s}^{-1}$ for configuration A, and $2.05 \times 10^{-5} \text{ mol m}^{-2} \text{ s}^{-1}$ for the reference configuration B ($\sigma = 3 \times 10^{-6} \text{ s}^{-1}$). These results are the CH_4 emission rates to the atmosphere for configurations A and B, respectively. The methane flux toward the atmosphere for the configuration without soil cover is twofold the one with a 0.5 m soil cover (configuration B ($\sigma = 3 \times 10^{-6} \text{ s}^{-1}$)).

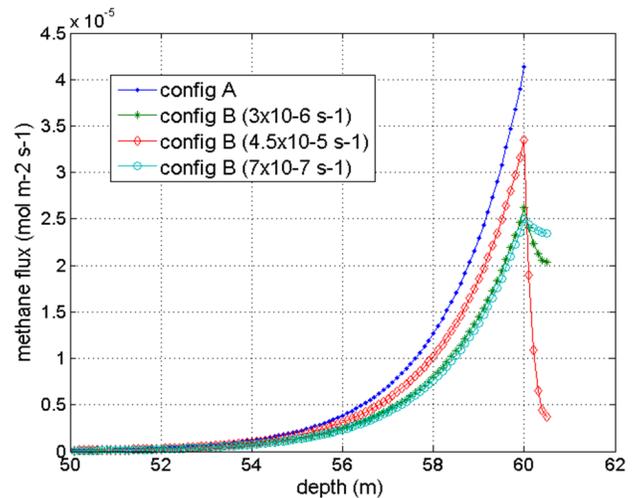


Fig. (4). Methane flux as function of depth for configuration A and configuration B for three different values of oxidation coefficient.

To consider the impact of varying oxidation coefficients on the results we also display in Fig. (4) the methane flux for small and large values of oxidation coefficient for the cover region: $7 \times 10^{-7} \text{ s}^{-1}$ and $4.5 \times 10^{-5} \text{ s}^{-1}$.

Methane Flux Measurements at the CTVM-Caieiras Site

The calculated results were compared with methane fluxes measured at 11 different locations in which the MSW were disposed during 2005 and 2007. Since the survey took place in 2010 it was considered an average waste age of 4 years. The influence of neighboring extraction wells could not be eliminated since the distance between wells was about 45 m. One of the measurements furnished a small negative flux indicating that there was possibly an influx of methane from the atmosphere to the landfill. Fig. (5) shows the methane flux at 10 locations where the measurement results yielded positive fluxes. The error in each measurement was estimated as 11 % [7]. The geospatial mean for the methane flux was $1.4 \pm 2.4 \times 10^{-4} \text{ mol m}^{-2} \text{ s}^{-1}$, the maximum was $6.7 \times 10^{-4} \text{ mol m}^{-2} \text{ s}^{-1}$, the minimum, $1.7 \times 10^{-6} \text{ mol m}^{-2} \text{ s}^{-1}$, and the median was $1.9 \times 10^{-5} \text{ mol m}^{-2} \text{ s}^{-1}$. Since there are very small and very large values of methane flux we arranged the data in Fig. (4) according to their magnitude.

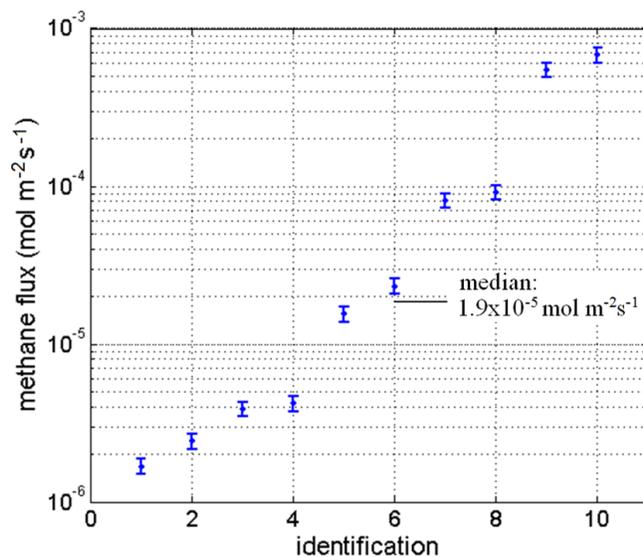


Fig. (5). Methane flux measurements at 10 different locations in the CTVM-Caieiras site.

DISCUSSIONS

Comparison Between Calculated and Measured Results

The calculated methane flux for the reference configuration B agreed well with the median value of the field measurements, but it was 7 fold smaller than its geospatial mean. The calculated results with small and large oxidation coefficients reproduced better the field measurements at locations with lower methane fluxes (identifications 3 to 6). Despite the good agreement between calculated methane flux to the atmosphere and the median of the experimental results presented in Fig. (5), one cannot say that this simplified approach is accurate. The experimental results spanned three orders of magnitude and thus specific conditions of methane emissions must be considered.

Table 2 presents field measurements in different landfills and the results obtained in this article [4,13,17,21-23]. Emission rates vary a lot in large landfills and usually field measurements are quoted as minimum and maximum values. The experimental median and the calculated methane fluxes compare well with the emission data reported by several authors [4,13,17,21], while the experimental geospatial mean compared better with the results of others [22,23]. Chanton *et al.* [4] and Abichout *et al.* [24] observe that spatial means of methane fluxes are usually dominated by “hotspots” with large emissions due possibly to the presence of macro-pores, preferential flow routes, different methane generation rates, and specific transport conditions. The data from Refs. 22 and 23 include landfill sections with thin soil covers, and such “hot spot” locations. The high methane fluxes of locations 9 and 10 in Fig. (5) could be considered due to such “hot spots” in the CTVM-Caieiras landfill.

The results of methane fluxes to the atmosphere are strongly dependent on the oxidation coefficient utilized for the cover region (see Fig. 4). Excluding “hot spot” emission conditions, the results evidence that the approach can reproduce any experimental value with adequate transport parameters. Since the methane concentration and flux near the atmosphere interface fall off as a combination of exponential functions (Table A3 in the Appendix), a value for the parameter $\beta_C = \sqrt{\sigma_C/D_C}$ can be obtained, representing a specific soil cover material and microclimate conditions, so that calculated results reproduce experimental results.

Modeling Lateral Migration to Extraction wells Through Transversal Sorption Coefficients

The methane concentration in the bottom of the landfill determines the gradient of the methane concentration across the upper part of the MSW and the cover regions, and thus the emission rate to the atmosphere. Extraction wells usually cause a transversal migration of methane, decrease its concentration in the bottom of the landfill and, consequently, its internal pressure there. This transversal migration in one-dimensional models is usually accounted for considering that only a fraction of the total generated methane moves toward the atmosphere [4,5,11]. Observing the analytical expressions in Table A3 for the methane concentration and flux we note that their magnitudes are controlled by the ratio R_0/σ_w ($\sigma_w = \sigma_T$ in the MSW region). Thus the influence of extraction wells could be modeled by assigning a value for the transversal sorption coefficient, σ_T , to adjust the internal pressure in the landfill bottom to actual values. In this work the transversal sorption coefficient was chosen to produce an internal pressure of 1 atm.

Methane Oxidation in the Cover Region

The oxidation coefficient was estimated from experimental results obtained in environmental conditions close to those of the CTVM-Caieiras site. Given the strong variation of oxygen concentration and population of methanotroph microorganisms in the cover region, and variable environmental conditions, a homogenous oxidation coefficient for the cover region appears to be much approximated. But the results allow qualitative analyses about the transport and oxidation phenomena. The results of

Table 2. Measured methane fluxes at various landfills and calculated results with different oxidation coefficients.

	Cover Thickness (m)	Methane Flux (mol m ⁻² s ⁻¹)
Ref. 22	0.2 - 0.7	2.8 x 10 ⁻⁴
Ref. 23	0.2 - 0.9	2.6 x 10 ⁻⁴
Ref. 21	1	2.4 x 10 ⁻⁵
Ref. 17	1	2.1 x 10 ⁻⁵
Ref. 13	0.8	4.2 x 10 ⁻⁵
Ref. 4 - Landfill X South	0.3 - 0.45	2.1x10 ⁻⁵
This work		
• (measured median)	0.5	2.0x10 ⁻⁵
• (measured geospatial mean)	0.5	1.4x10 ⁻⁴
• "A" configuration (calculated)	0	4.2 x10 ⁻⁵
• $\sigma = 7 \times 10^{-7} \text{ s}^{-1}$ (calculated)	0.5	2.3x10 ⁻⁵
• $\sigma = 3 \times 10^{-6} \text{ s}^{-1}$ (calculated)	0.5	2.0x10 ⁻⁵
• $\sigma = 4.5 \times 10^{-5} \text{ s}^{-1}$ (calculated)	0.5	3.5x10 ⁻⁶

Figs. (2, 3) show that the oxidation coefficient for the cover region, σ_{ox} , should lie between $7 \times 10^{-7} \text{ s}^{-1}$ and $4.5 \times 10^{-5} \text{ s}^{-1}$. In the range from $7 \times 10^{-7} \text{ s}^{-1}$ to $3 \times 10^{-6} \text{ s}^{-1}$ σ_{ox} represents weak oxidation conditions since the methane flux to the atmosphere decreased only 15 %, while from $3 \times 10^{-6} \text{ s}^{-1}$ to $4.5 \times 10^{-5} \text{ s}^{-1}$ represents stronger oxidation conditions since the methane flux decreased about 90 %. Values of σ_{ox} larger than 10^{-4} s^{-1} represent conditions of very strong methane oxidation and decrease the methane flux to the atmosphere to negligible values.

The results of Fig. (4) and Table A3 indicate that the methane oxidation can be increased by either increasing the oxidation capability of the cover soil (having greater β_c by increasing the soil σ_{ox}) or increasing the cover soil thickness. Apart the approximations considered in this article, it illustrates how less effective soil material utilized in landfill covers can be compensated by thicker soil covers to reduce methane emission.

Influence of Waste Characteristics

For covered landfills the important waste characteristic is its methane generation capacity characterized by the parameter R_0 . The oxidation rate in this region is low or negligible due to lack of oxygen and greater in the cover region. For such landfill configuration, and with active extraction wells, the dominant effect in the waste region is the transversal migration toward these wells. For landfills without cover region, the MSW dispersion coefficient determines the emission rate.

CONCLUSION

The analytical expressions developed in this article allowed obtaining estimates for methane emissions from landfills to the atmosphere, taking into account the oxidation that occurs in the cover region. The introduction of an explicit oxidation coefficient and the assumption of homogenous conditions yielded analytical solutions for one-dimensional emission

problems. The methane lateral migration toward extraction wells was treated as a fictitious transversal methane sorption. The obtained results presented good agreement with the median of measured methane fluxes conducted at the CVTM-Caieiras site. Similarly to other calculation schemes for methane emission, good comparisons with experimental results depend on the parameters considered to describe the problem which varies with site and microclimate conditions (in this work, the methane oxidation coefficient).

The analytical expression revealed qualitative description of the transport phenomena rather than accurate results for all conditions. For instance, the usual observation of methane concentration decay near the atmosphere interface is described by a combination of exponentials characterized by the oxidation coefficient. This parameter can be considered a soil cover property related to the probability per unit of time that the methane is oxidized in the cover region. In principle it depends on the type of soil used and environmental conditions such as pH, temperature, moisture and the concentrations of O₂ and other gases. Since the methane concentration falls off as a combination of exponential functions, the parameter $\beta_c = \sqrt{\sigma_c/D_c}$ can be measured for specific soils, and microclimate conditions through the fitting of the analytical solution for the methane concentration to experimental methane concentration profiles.

CONFLICT OF INTEREST

The authors confirm that this article content has no conflict of interest.

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APPENDIX

Table A1. Waste characteristics of the CTVA-Caieiras landfill [7].

Parameter*	Value
MSW porosity	0.30
Soil cover porosity	0.22
Biodegradation constant for the MSW, k	0.09 year ⁻¹
Methane generation potential of MSW, L	4.15x10 ⁻³ mol kg ⁻¹
Specific mass of MSW	600 kg m ⁻³
MSW average age	4 years
Uniform methane generation rate, R ₀	2.45x10 ⁻⁵ mol m ⁻³ s ⁻¹

Table A2. Parameters describing the CTVA-Caieiras landfill.

Parameter*	Value
Methane diffusion coefficient in the air [20]	2.12x10 ⁻⁵ m ² s ⁻¹
c parameter [5]	0.15
d parameter [5]	1.1
γ factor for the MSW material	0.113
γ factor for the soil cover material	0.075
D _w dispersion coefficient in the MSW	3.14x10 ⁻⁶ m ² s ⁻¹
D _C dispersion coefficient in the soil cover	1.36x10 ⁻⁶ m ² s ⁻¹
σ _T - transversal sorption coefficient in the MSW region	1.1x10 ⁻⁶ s ⁻¹
σ _{ox} - oxidation sorption coefficient	3x10 ⁻⁶ s ⁻¹
K _c - half saturation concentration for CH ₄	0.2 mol m ⁻³
K _o - half saturation concentration for O ₂	0.4 mol m ⁻³
V _m - maximum methane oxidation	3.36x10 ⁻⁵ mol m ⁻³ s ⁻¹
Reference oxygen concentration	4 mol m ⁻³

*Subscripts w and c mean MSW and soil cover regions, respectively. More information can be found in Ref. [7].

Table A3. Expressions obtained from the analytical solution of the methane transport equation for configurations A and B.

<p>Methane concentration for configuration A</p> $C(z) = \frac{R_0}{\sigma_w} \left(1 - \frac{\cosh(\beta_w z)}{\cosh(\beta_w H)} \right)$
<p>Methane concentration for configuration B</p> $C(z) = \begin{cases} \frac{R_0}{\sigma_w} (1 - A_w \cosh(\beta_w z)), & \text{for } 0 \leq z \leq H, \text{ (MSW region)} \\ \frac{R_0}{\sigma_w} A_c \sinh(\beta_c(L - z)), & \text{for } H \leq z \leq L, \text{ (cover region)} \end{cases}$
<p>Methane flux for configuration B</p> $J(z) = \begin{cases} \frac{R_0}{\sigma_w} A_w D_w \beta_w \sinh(\beta_w z), & \text{for } 0 \leq z \leq H \text{ (MSW region)} \\ \frac{R_0}{\sigma_w} A_c D_c \beta_c \cosh(\beta_c(L - z)), & \text{for } H \leq z \leq L \text{ (cover region)} \end{cases}$
$\beta_w = \sqrt{\sigma_w/D_w}, \beta_c = \sqrt{\sigma_c/D_c}, F_{wc} = \frac{D_w \beta_w \sinh(\beta_w H)}{D_c \beta_c \cosh(\beta_c(L-H))}$ $A_w = \frac{1}{F_{wc} \sinh(\beta_c(L-H)) + \cosh(\beta_w H)}, A_c = A_w F_{wc}$

REFERENCES

- [1] V. B. Stein, J. P. A. Hettiaratchi, and G. Achari, "Numerical model for biological oxidation and migration of methane in soils", *Practice Periodical of Hazardous, Toxic, and Radioactive Waste Management*, vol. 5, pp. 225-234, 2001.
- [2] A. De Visscher and O. Van Cleemput, "Simulation model for gas diffusion and methane oxidation in landfill cover soils", *Waste Management*, vol. 23, no.7, pp. 581-591, 2003.
- [3] K. A. Spokas and J. E. Bogner, "Limits and dynamics of methane oxidation in landfill cover soils", *Waste Management*, vol. 31, no. 5, pp. 823-832, 2011.
- [4] J. Chanton, T. Abichou, C. Langford, K. Spokas, G. Hater, R. Green, D. Goldsmith and M. A. Barlaz, "Observations on the methane oxidation capacity of landfill soils", *Waste Management*, vol. 31, no. 3, pp. 914-925, 2011.
- [5] M. D. N. Perera, J. P. A. Hettiaratchi and G. Achari, "A mathematical modeling approach to improve the point estimation of landfill gas surface emissions using the flux chamber technique", *Journal of Environmental Engineering and Science*, vol. 1, no. 6, pp. 451-463, 2002.
- [6] V. C. Hettiarachchi, J. P. A. Hettiaratchi and A. K. Mehrotra, "Comprehensive one-dimensional mathematical model for heat, gas, and moisture transport in methane biofilters", *Waste Management*, vol. 11, no. 4, pp. 226-233, 2007.
- [7] G. Candiani, "Estudo da geração de metano em uma célula de aterro sanitário", Ph. D. thesis, Universidade Federal do ABC, Santo André, SP, Brazil, 2011.
- [8] G. Tchobanoglous, H. Theisen and S. A. Vigil, *Integrated solid waste management - engineering principles and management issues*, McGraw-Hill: NY 1993.
- [9] C. K. Wang and G. Achari, "A comparison of performance of landfill gas collection wells with varying configurations", *The Open Waste Management Journal*, vol. 5, no. 3, pp. 40-48, 2012.
- [10] J. Peischl, T. B. Ryerson, J. Brioude, K. C. Aikin, A. E. Andrews, E. Atlas, D. Blake, B. C. Daube, J. A. Gouw, E. Dlugokencky, G. J. Frost, D. R. Gentner, J. B. Gilman, A. H. Goldstein, R. A. Harley, J. S. Holloway, J. Kofler, W. C. Kuster, P. M. Lang, P. C. Novelli, G. W. Santoni, M. Trainer, S. C. Wofsy and D. D. Parrish, "Quantifying sources of methane using light alkanes in the Los Angeles basin, California", *Journal of Geophysical Research: Atmospheres*, vol. 118, pp. 4974-4990, 2013.
- [11] L. A. K. Perera, G. Achari and J. P. A. Hettiaratchi, "Determination of source strength of landfill gas: a numerical modeling approach", *Journal of Environmental Engineering*, vol. 128, no. 5, pp. 461-471, 2002.
- [12] J. Im, S. Moon, K. Nam, Y. J. Kim and J. Y. Kim, "Estimation of mass transport parameters of gases for quantifying CH₄ oxidation in landfill soil covers", *Waste Management*, vol. 29, no. 7, pp. 869-875, 2009.
- [13] T. Abichou, K. Mathieu, L. Yuan, J. Chanton and G. Hater, "Effects of compost biocovers on gas flow and methane oxidation in a landfill cover", *Waste Management*, vol. 29, no.5, pp. 1595-1601, 2009.
- [14] E. R. Silva, "Modelagem matemática da produção e transporte de biogás em aterros sanitários", M. S. thesis, Universidade Federal do ABC, Santo André, SP, Brazil, 2010.
- [15] J. Gebert, A. Groengroeft and G. Miehlich, "Kinetics of microbial landfill methane oxidation in biofilters", *Waste Management*, vol. 23, no.5, pp. 609-619, 2003.
- [16] M. Pawlowski and W. Stepniwski, "Biochemical reduction of methane emission from landfills", *Environmental Engineering Science*, vol. 23, no.4, pp. 666-672, 2006.
- [17] C. Scheutz, J. Bogner, J. P. Chanton, D. Blake, M. Morcet, C. Aran and P. Kjeldsen, "Atmospheric emissions and attenuations of non-methane organic compounds in cover soils at a French landfill", *Waste Management*, vol. 28, no.6, 1892-1908, 2008.
- [18] C. Scheutz and P. Kjeldsen, "Environmental factors influencing attenuation of methane and hydrochlorofluorocarbons in landfill cover soils", *Journal of Environmental Quality*, vol. 33, pp. 72-79, 2004.
- [19] C. Scheutz, P. Kjeldsen, J. E. Bogner, A. De Visscher, J. Gebert, H. A. Hilger, M. Huber-Humer and K. Spokas, "Microbial methane oxidation processes and technologies for mitigation of landfill gas emissions", *Waste Management & Research*, vol. 27, pp. 409-455, 2009.

- [20] A. De Visscher, D. Thomas, P. Boeckx and O. Van Cleemput, "Methane oxidation in simulated landfill cover soil environment", *Environmental Science & Technology*, vol. 33, no.11, pp. 1854-1859, 1999.
- [21] M. Christophersen, H. Holst, J. Chanton and P. Kjeldsen, "Lateral gas transport in soil adjacent to an old landfill: factors governing emission and methane oxidation", *Waste Management and Research*, vol. 19, pp. 595-612, 2001.
- [22] F. J. Maciel, "*Estudo da geração, percolação e emissão de gases no aterro de resíduos sólidos na Muribeca/PE*", M. S. thesis, Universidade Federal de Pernambuco, Recife, PE, Brazil, 2003.
- [23] M. O. H. Mariano, "*Avaliação da retenção de gases em camadas de cobertura de aterros de resíduos*", M. S. thesis, Universidade Federal de Pernambuco, Recife, PE, Brazil, 2008.
- [24] T. Abichou, D. Powelson, J. Chanton, S. Escoriza and J. Stern, "Characterization of methane flux and oxidation at a solid waste landfill", *Journal of Environmental Engineering*, vol. 132, no.11, pp. 220-228, 2006.

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