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APPLICATION OF PERCOLATION THEORY AND FRACTAL GEOMETRY TO LANDFILL METHANE PRODUCTION

APLICACIÓN DE LA TEORÍA DE PERCOLACIÓN Y LA GEOMETRÍA FRACTAL EN LA PRODUCCIÓN DE METANO EN RELLENO SANITARIO

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Abstract

The estimation of methane generation in a landfill is based mainly on first-order chemical kinetics, which is a good theory for gaseous media or in solution, i.e. a homogeneous media; however in a landfill, conversions lower than expected yields are observed. In this paper, we propose the consideration of the sanitary landfill heterogeneity, through the classic percolation theory, and introducing a β factor in first order models to reduce the error of the methane estimations in the landfill. This percolation factor (β) only considers the degradation of organic matter in methanogenic zones, which we propose immersed in the percolation clusters (including the infinite cluster), so $\beta = \{1/4, 2/3\}$. When including β in the Hoeks model (Hoeks, 1983), and fractal-like Hoeks model (Meraz *et al*., 2004), conversions of 22-59% and 12-33% are reached at 30 years of closed site, respectively. These ranges are within what was estimated by Bogner and Spokas (1993), 25-40% conversion for the same time period.

Keywords: classical percolation, landfill, methane, fractal-like chemical kinetics, modeling.

Resumen

La estimación de la generación de metano en relleno sanitario está basada principalmente en cinéticas químicas de primer orden, que es una teoría acertada para medios gaseosos o en solución, i.e. medios homogéneos. Sin embargo, en el relleno sanitario, se observan conversiones menores a las predichas. En este trabajo proponemos considerar la heterogeneidad del relleno sanitario a través de la teoría de percolación clásica y se introduce el factor β en modelos de primer orden para reducir el error de las estimaciones de metano en el sitio. Este factor de percolación (β) sólo considera la degradación de la materia orgánica en zonas metanógenas, las cuales proponemos inmersas en los racimos de percolación (incluido el racimo de percolación infinito) y así, $\beta = \{1/4, 2/3\}$. Al incluir β en los modelos de Hoeks (1983) y en el modelo Hoeks fractal (Meraz *et al*., 2004), las conversiones son 22-59% y 12-33%, respectivamente, a 30 años de cerrado el sitio. Estos intervalos se encuentran dentro de lo estimado por Bogner y Spokas (1993), 25-40% de conversión dentro del mismo periodo.

Palabras clave: percolación clásica, relleno sanitario, metano, cinética química fractal, modelación.

1 Introduction

Landfilling of municipal solid waste implies the anaerobic digestion of biodegradable material to generate biogas. Methane, which constitutes about 55% of biogas volume, is the major interest constituent for sizing the gas collection systems of landfills, for evaluation of the benefits of gas recovery projects (Aldana-Espitia *et al*., 2017) and estimating gaseous emissions to the atmosphere. Hence the two most important components of model predictions are the amount of methane that can ultimately be generated from a given amount of garbage buried in the landfill, and the rate of generation in the course of time (Tchobanoglous and Theisen, 1993). Currently, the First Order Decay empirical methods (FOD methods) are very used in the landfill for methane production estimation (Amini *et al*., 2012; Amini *et al*., 2013; Chakraborty *et al*., 2011; Mønster *et al*, 2015; Hoeks, 1983; Thompson *et al*., 2009). The FOD methods are based on a chemical reaction of the type $A \rightarrow X$ (we take the simplest case for didactic reasons). The degradation rate of the reagent is $dA/dt = -k[A]$, and for the generation of the product results $dX/dt = k[A]$. The solution to these first-order differential equations are $A = A_0 \exp(-kt)$, and $X = A_0[1 - \exp(-kt)]$. Note that in this solution, by knowing the initial reagent A_0 amount, the quantity formed of the X product can be

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known at any t_f time. However, in the landfill, this relation is somewhat different (Krause *et al*., 2016a).

A landfill is a bad reactor because its operation implies highly compaction, with low water content and imperfect mixing (*cf*. Tchobanoglous and Theisen, 1993). In these conditions, the accumulated experience shows that landfill stabilization is extremely slow and it is common to detect well-conserved waste decades after site closure (Bautista-Ramírez *et al*., 2018; Li *et al*., 2009). Bogner and Spokas (1993) found that the conversion is less than 40% thirty years after landfill closure. Furthermore, gas production has been observed 100 years after site closure (Finnveden and Nielsen, 1999). Thus, in the field, classical chemical concepts are not enough to answer important questions surrounding the generation of methane in the landfill. Such a scenario requires to incorporate concepts of complex and disordered systems (*cf*. Ben-Abraham and Havlin, 2004; Sahimi, 1993) to the modeling of methane production in landfill (in this work we will only focus on this biogas component), such as the percolation theory and fractal geometry, for a better understanding of the relationship between methane generation and waste heterogeneity.

This manuscript presents the material as follows. First, the model framework: i) the reaction scenario is described, and considered as a system of heterogeneous nature without agitation; ii) a selection of useful concepts of classical percolation theory is provided, which allows for establishing a factor that implies the real organic matter fraction available in the MSW bulk to generate methane; iii) Hoeks model, i.e. a FOD method, and fractal-like Hoeks model are outlined, and this last is modified with the percolation factor; and, iv) the methane potential generation calculation by a global chemistry reaction is showed. Finally, using a numerical study with a hypothetical landfill proposed, the resulting methane productions by modified FOD method equation are compared to field estimations. We can see a significant reduction of methane production estimations by using the percolation factor and fractal geometry.

2 Methodology

2.1 Reaction medium geometry

A landfill involves engineering methods for waste disposal and special handling to control environmental impacts. Landfill operation is based on the daily placement of waste in a specified area called a cell, whose background is isolated from the environment through a synthetic membrane. In such conditions, there is poor mixing and chemical reactions are controlled by diffusion through a heterogeneous medium. Residues can be divided into four main kinetics categories: i) readily biodegradable refuse fraction (food), which is susceptible to fast acidogenesis (De la Cruz and Barlaz, 2010; Eleazer *et al*., 1997; Fei *et al*., 2016); ii) moderately biodegradable refuse fraction (garden waste). Food and garden waste mainly determine the bacteria population strength and the nutrient availability in the site (Barlaz, 2000); iii) slowly biodegradable refuse fraction (paper, wood and textiles), which is the main methane source. It is known that paper waste is the foremost methane producer. Thus recycling of this raw material should have major effects on landfill methane yield (Aranda *et al*., 2008); and iv) inert materials, usually impermeable materials that determine the medium tortuosity (Sanchez *et al*., 2007). Thus, the reaction medium is formed by permeable and impermeable elements. The permeable zone provides the necessary moisture for the substrate diffusion and transport of methanogenic organisms (Stanley *et al*., 2011).

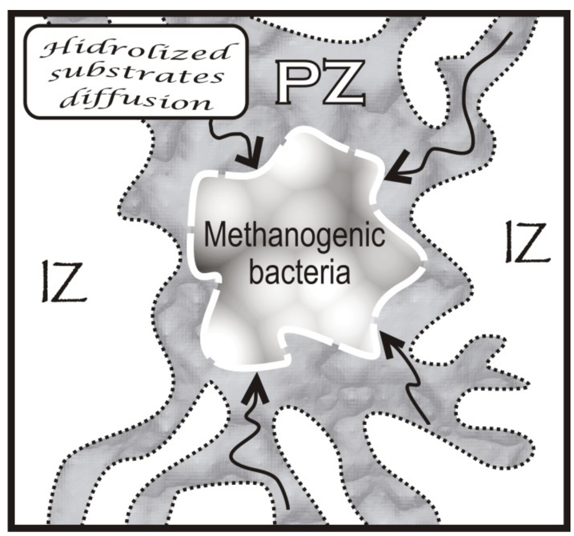

Fig. 1. A dual distributed model of methanogenesis in landfill. There are two different zones in the volume occupied by residues: i) a permeable zone (PZ) where methanogens are safe with sufficiently moisture, and consume the hydrolyzed substrates (acetic acid molecules are transported by diffusion) to produce methane, and ii) an impermeable zone (IZ) with inert and trapped organic waste.

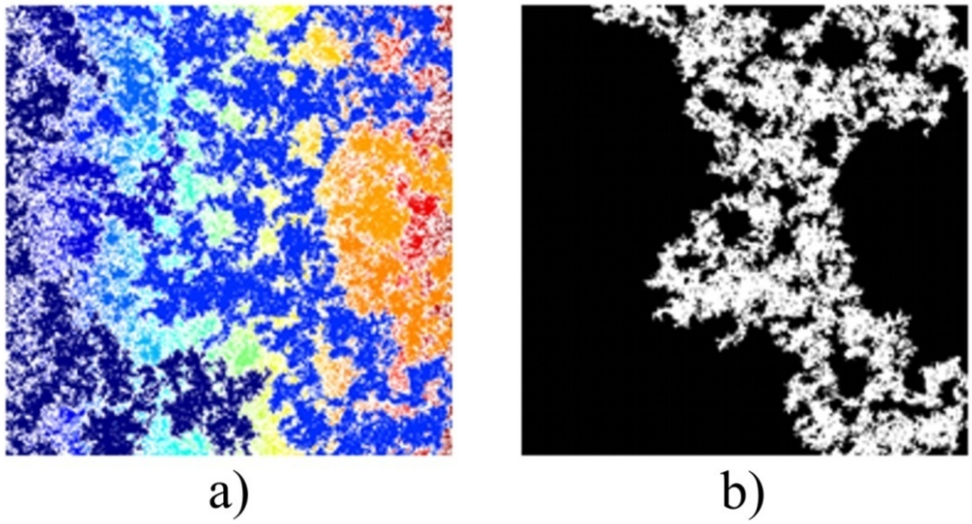

Fig. 2. Classical percolation on 1000 x 1000 sites network. Network boundaries are closed to any type of fluid flow with the sole exception of two producing sites located at opposite edges (up and down) of the network. a) An illustration of the population of clusters at percolation threshold. The different clusters are presented with different color; the infinite cluster is light blue. b) Black and white representation of the cluster, the permeable fraction is depicted in white and the impermeable one in black.

On the other hand, the impermeable zone prevents the inhibition of methanogenic zones by acidogenesis products (Martin *et al*., 1997), although this zone can include trapped organic refuse, see Fig. 1. The theory of percolation is developed under a dual idea (occupied or not), so to conceptualize the environment as a permeable zone or not, this theory can be taken as explained below.

2.2 *Percolation theory and landfill*

Landfill heterogeneity is due to the varied nature of waste (Zacharof and Butler, 2004). This heterogeneity is well represented through the application of stochastic models such as classical percolation theory (Isichenko, 1992; Sahimi, 1993; Stauffer, 1985), which is based on the movement of a fluid in a random medium (Daccord, 1987; Lenormand and Daccord, 1988). This medium is represented by a numerical matrix called a network, composed of two elements of different categories: the sites and the bonds. In analogy with a porous medium, the bonds must be narrower than the sites (Mayagoitia *et al*., 1989; Riccardo *et al*., 1993). When the fluid manages to cross the network, it is said that an infinite cluster of percolation has been formed with a fraction ρ_C of the sites called the classical percolation threshold. Depending on the number of bonds that connect to a site, different conformations of the network can be obtained. In the particular case of site percolation on random square networks, if one site connected with four bonds, ρ_C = 0.593 (Ben-Avraham and Havlin, 2004). Note that more than 60% (in mass) of organic matter of waste content guarantee percolation on a random square network. During the growth of the infinite cluster of percolation, also the fluid fills other zones (other clusters) that are not part of the infinite cluster. Figure 2 provides an illustration of both the clusters size distribution and the infinite cluster. The spanning site percolation cluster is shown in light blue besides other smaller clusters. Note the complex morphology of the percolating cluster including a convoluted boundary and 'holes' of various sizes inside the cluster. Percolation models present highly tortuous paths and significant 'dead-end' zones. Hidalgo-Olguín *et al*. (2015) found that the fraction occupied by the infinite percolation cluster φ for random square networks is ≈0.25, that is $\varphi < \rho_C$.

The fraction of biodegradable matter in waste (*cf*. Table 1) is somewhat greater than the percolation threshold for square lattice (ρ_C = 0.593), thus φ can easily reach a value of ~2/3. Then, if it is considered that in the landfill the permeable and reactive zones (Staley *et al*., 2011) are the occupied zones in classic percolation, then the largest generation of methane can be attributed to a quarter of the organic matter deposited, because of $\varphi \approx 1/4$.

Table 1 Typical Mexican waste composition. Its biodegradability, composition, water content, and potential methane generation by tonne of MSW as discarded, under standard conditions. Composition and biodegradability are taken from Meraz *et al*. (2004). Methane generation is computed via Eq. (4).

Waste	Biodegradability	%Mass	%Water content	Methane generation by tonne of MSW as discarded (m^3/tonne)
Food	Readily	39	70	40.233
Garden	Moderately	6	60	8.253
Paper	Slowly	22	6	71.112
Textiles	Slowly	2	10	6.19
Wood	Slowly	1	20	2.751
Plastics	Inert	6	2	-
Glass	Inert	8	2	-
Metal	Inert	4	3	-
Ash, rock, and dirt	Inert	12	8	-

Meaning that the organic matter that allows the methane generation is in the infinite cluster of percolation, although it is not necessary to discard that other permeable, and reactive zones exist when $\varphi \approx 2/3$. This factor henceforth named the percolation factor β, can be introduced in first-order models such as Hoeks, for wich $\beta = \{1/4, 2/3\}$.

2.3 *Modifications of the Hoeks model*

In the Hoeks model (1983), a FOD method, the MSW is partitioned in four kinetics categories (see Section 2.1): readily, moderately, and slowly biodegradable residues, and inert materials. Then, three categories of organic matter with different chemical kinetics behaviors are considered in the model:

$$Q_{CH_4} = L_0 \sum_j \sum_i M_j C^0_{ij} k_i \exp(-k_i t_j) \quad (1)$$

where M_j is the mass waste buried in j year, C^0_{ij} is the fraction of dry organic matter mass of kind i in j year, k_i is the rate constant of organic fraction i, and t_j is the year j. Each kind of organic matter is associated with a first-order kinetic rate constant determined by Hoeks under good reaction conditions (*i.e*., a mixed system with enough water, pH control, etc.) as: $k_1 = 0.69 \pm 0.23$ year^{-1}, $k_2 = 0.14 \pm 0.01$ year^{-1}, $k_3 = 0.05 \pm 0.02$year^{-1}, for readily, moderately, and slowly biodegradable residues, respectively. The half-life time values were estimated too: i) ~ 1 year for readily biodegradable residues, ii) ~5 years for moderately biodegradable residues, and iii) ~15 years for slowly biodegradable residues.

The fractal-like chemical kinetics (Kopelman, 1988) is another way to approach a complex medium such as landfill, with inherent and ubiquitous irregularity, where kinetics is controlled by diffusion in a convoluted and not mixed environment, and is difficult for conventional theories to address (Pensin and Climenhaga, 2009; Rothschild, 1998). A modification to Hoeks equation was introduced in Meraz *et al*. (2004) by considering fractal-like chemical kinetics, leading to:

$$Q_{CH_4} = L_0 \sum_j \sum_i M_j C^0_{ij} k_i \exp(-k_i t_j) t_j^{-h} \quad (2)$$

Note that when $h = 0$, Eq. (2) reduces to Eq. (1), it means that there is a homogeneous medium, but there is not a heterogeneous medium (fractal-like medium). In this last case, $0 < h \leq 1$, i.e. in a tortuous and poorly stirred medium. The Alexander-Orbach conjecture sets $d_s = 4/3$ (spectral dimensionality) as the most likely value for a fractal medium (Alexander and Orbach, 1982; Havlin and Ben-Avraham, 2002; Kozma and Nachmias, 2009). In consequence, if $h = 1 - d_s/2$ (Kopelman, 1988), then $h = 1/3$. Thus, the parameter h directly affects the rate constant k and implies the decrease of the methane production (Kopelman, 1989).

Finally, the inclusion of β in Eq. (2) [and in the same manner in Eq. (1)], results in

$$Q_{CH_4} = \beta L_0 \sum_j \sum_i M_j C^0_{ij} k_i \exp(-k_i t_j) t_j^{-h} \quad (3)$$

2.4 *Methane potential generation*

The microbiological decomposition of the organic fraction contained in the waste deposited at the site produces biogas and can be identified with Batch

Solid-State Digestion (BSSD) (Ko *et al*., 2015; Martin, 2001; Xu *et al*., 2015). It is known that degradation of organic waste buried in landfill passes through a typical three-phase process (Tchobanoglous and Theisen, 1993): i) *Aerobic*. This phase leads to oxygen exhaustion in the fill due to a short aerobic activity (weeks), that is irrelevant with regard to methane production and is not considered in this work. The following two phases are essential parts of our model given their importance in methane generation. ii) *Acidogenesis*. The large organic molecules in the organic waste are broken down by polymer hydrolysis leading to the creation of acetic acid (Franke-Whittle *et al*., 2014). This phase is catalyzed by a community of bacteria (acidogenic bacteria). iii) *Methanogenesis*. This is the longest period which lasts for decades. Here, methane is produced by methanogenic bacteria (Vavilin and Angelidaki, 2005). With the idea that glucose composition is representative of all organic matter in waste, and considering NH_3 as the nitrogen source, the following global material balance for organic anaerobic degradation has been established (Meraz and Domínguez, 1998),

$$C_6H_{12}O_{6(s)} + 0.18NH_{3(g)} \rightarrow 2.53CH_{4(g)} + 2.54CO_{2(g)} + 0.42H_2O_{(l)} + 0.93CH_{1.7}O_{0.5}N_{0.2(s)} \tag{4}$$

Eq. (4) states the following: i) the generation of biogas, a gas mixture with a significant energetic value due to its methane content, ii) the production of bacterial cell mass ($CH_{1.7}O_{0.5}N_{0.2}$) and the by-products of bacterial metabolism (CO_2, CH_4, and H_2O) when anaerobic bacteria consume the biodegradable organic matter ($C_6H_{12}O_6$). Note that Eq. (4) is a global reaction that explicitly considers the generation of biomass. This is an advantage over global reactions based on the stoichiometric content of carbon only (Krause *et al*., 2016b). Besides, Eq. (4) represents an interesting option with respect to the methodology used in the field to estimate the methane potential of the waste, which usually uses firsthand values and empirical formulas (Krause *et al*., 2016a). Using the typical Mexican waste composition presented in Table 1, and by means of Eq. (4), the methane generation (m^3) under standard conditions, by a tonne of municipal solid waste (MSW) as discarded, of every biodegradable MSW component is calculated and given in the last column of Table 1. It is pertinent to mention that the methane generation values given in Table 1 are never observed in landfills over a period of 30 years from the beginning of site activities (Bogner and Spokas, 1993), due to transport difficulties and acid inhibition.

3 Results and discussion

In order to compare the methane production versus time curves predicted by the different equations, a virtual study is developed on a hypothetical site that has been landfilled for 9 years. The mass dumped over this period is 7500 ktonne. From data in Table 1, and using Eq. (4), it is easily shown that for typical Mexican residues composition, L_0 = 128.538 m^3/tonne under standard conditions. Figure 3 shows the behavior of Hoeks model [Eq. (1)], Hoeks fractal-like equation with $h = 1/3$, Eq. (2), and Eq. (3) with $h = 1/3$ and $\beta = \{1/4, 2/3\}$.The calculations were realized with Excel software.

Note that all equations give a maximum of methane production that corresponds to landfill completion. Then a monotonous decay stage of methane production follows, and 50 years after site closure a significant methane production is still observed. The peak of methane production is significantly diminished by the medium fractality. The value of the percolation factor β further contracts the peak height.

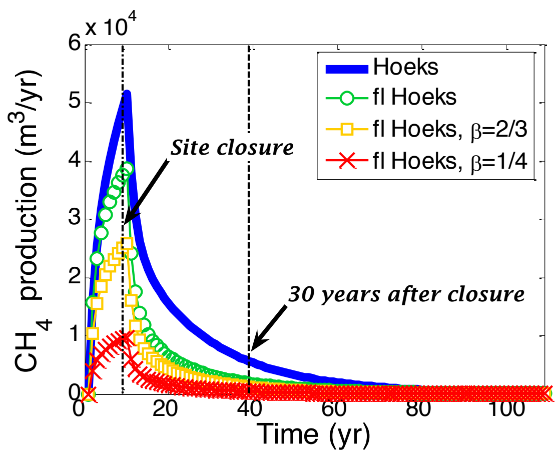

Fig. 3. Comparison between methane production prediction from Hoeks model (continues line), Eq. (1), ($h = 0, \beta = 1$), fractal-like Hoeks model (fl Hoeks, line with circles), Eq. (2), ($h = 1/3$, $\beta = 1$), and fractal-like Hoeks model with percolation effect, Eq. (3), with $h = 1/3$ and $\beta = \{1/4, 2/3\}$ (lines with squares and crosses, respectively).

Table 2 Conversion at 0, 14, 30, 50 and 100 years for a hypothetical site closure. This site is formed by 9 layers. Data from the application of Hoeks equation, Eq. (1), ($h = 0, \beta = 1$), the fractal-like Hoeks equation, Eq. (2), ($h = 1/3, \beta = 1$), and with percolation effect, Eq. (3), with $h = 1/3, \beta = \{1/4, 2/3\}$. % conversion = [cumulative methane production / amount of methane produced according to Eq. (4)] × 100.

Eq.	h	β	% conversion				
			0 yr	14 yr	30 yr	50 yr	100 yr
1	0	1	38	74	88	96	100
2	1/3	1	25	42	47	49	50
3	1/3	2/3	16	28	32	33	34
3	1/3	1/4	6	11	12	12	13

Table 2 presents some yield values for the different equations. We only considered the mean value of each kinetic rate constant for organic fraction of MSW.

Note that despite the uncertainty associated with kinetic constants, the differences between the diverse equations are clear. It is important to mention that during the landfilling, there is a 0 to 9 years lapse, equations (1) and (2) show that more than 1/4 of the methane potential yield of the site is lost to the atmosphere. In fact, methane cannot be efficiently recovered in this stage because of the gas recovery system is usually does not work in this period. Meanwhile, Eq. (3) predicts losses lower than 16 % for the same period. After site closure, methane production presents a monotonous decay for all equations, see Fig. 2. The half-life of slowly biodegradable waste is ∼14 years. Equation (1), considering a homogeneous reaction medium, indicates a very large conversion of 74% due to the presence in the fill of easily biodegradable waste. On the other hand, Eq. (2), taking into account the mass transfer difficulties imposed by the medium tortuosity, and lack of mixing, predicts a conversion slightly less than 50% (42%). Equation (3), which restrains the methane production to the infinite percolation cluster, significantly reduces the conversion of organic matter to methane. In the percolation threshold, $\beta = 2/3$ envisages a conversion of 28%, while Eq. (3) with $\beta = 1/4$, a low yield of 11% is predicted. The same trend is observed at 30 years from site closure. Equations (1) and (2) give conversions of 88% and 47%, respectively, larger than the 25-40 % yields that have been reported (*e.g.* Bogner and Spokas, 1993) at 30 years from site closure. Equation (3) gives conversions of 32% and 12%, for $\beta = 2/3$, and $\beta = 1/4$, respectively. These last values fall well within the reported values from field studies. It is convenient tosay that in the case of Hoeks model [Eq. (1)], the inclusion of factor β can changes the conversion from 88% at 30 years from site closure to 22 and 59% in the same period of time for $\beta = \{1/4, 2/3\}$, respectively. After, the predictions from all equations suggest that a significant methane production is still observed 50 years after site closure. Note that Eq. (1) shows that the reaction is near to finish (96%) at 50 years after site closure. In contrast, Eq. (3) indicates that just 12% of the site methane potential has been produced at 50 years from site closure for $\beta = 1/4$. Thus, we can say that for Eqs. (2) and (3) a steady-state is reached at 30 years from site closure. Even though, Eq. (3) with $\beta = 1/4$ achieved this state from 14 years. Finally, at 100 years after site closure, Eq. (3) with $\beta = 1/4$, shows that ∼10% of the site methane potential has been produced.

Conclusions

At least 60% of biodegradable material concentration in residues allows that a landfill is perceived as a percolative system, formed by clusters of the biodegradable and permeable material, slightly beyond the percolation threshold. Methane generation requires the establishment of a balance between the mechanisms of acidogenesis and methanogenesis, both occurring simultaneously in each cluster. Only the infinite cluster, being much larger than the others, provides sufficient connected space to establish such a balance, thus avoiding the accumulation of acids and the survival of methanogenic bacteria. Therefore, only the biodegradable material contained in the infinite cluster will be capable of producing methane. The rest of biodegradable material remains in vulnerable areas for inhibition by acidic species. In this work, a generic factor (β) based on the classical percolation concept has been proposed in order to estimate methane production from a landfill. Using the percolation factor β in Hoeks model [Eq. (1)] a FOD method, it is possible predicts at 30 years from site closure a conversion of 22-59%. The combination of two parameters of heterogeneity: β and fractality [Eq. (3)], point out a conversion of 10-30%, approximately, in the same period. Consequently, after 30 years the landfill can conserve between 40 and 90% of carbon as organic matter. The observations in the field show that the landfill is not an ideal reaction medium, so we believe that this work is useful to explain the discrepancies between the observations, and the estimates obtained by the FOD methods, through the use of models developed by unconventional methods, such as the percolation theory, in this area of environmental engineering.

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Nomenclature

A	reagent
A_0	initial concentration of reagent
C_{ij}^0	initial concentration of waste category i buried in year j
d_S	spectral dimensionality
h	Kopelman exponent
K	Kopelman kinetics coefficient
k	classic kinetics constant
k_1	first order chemical kinetics constant of readily biodegradable waste, yr^{-1}
k_2	first order chemical kinetics constant of moderately biodegradable waste, yr^{-1}
k_3	first order chemical kinetics constant of slowly biodegradable waste, yr^{-1}
k_i	first order chemical kinetics constant of organic matter of type i
L_0	potential methane yield of MSW, m^3/tonne under standard conditions
M_j	mass of waste landfilled during year j, kg
p	probability percolation parameter
P_∞	infinite cluster
Q_{CH_4}	methane generation rate, m^3/year under standard conditions
t	time, year
X	product
Greek symbols	
β	percolation factor
ρ_C	percolation threshold
φ	fraction occupied by the infinite percolation cluster

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