Using a Modeling Approach to Estimate Methane Emissions and Oxidation from Landfills Within AP-42 Guidelines.

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ABSTRACT: To estimate uncontrolled surface methane emissions from landfill surfaces, AP-42 guidelines are typically used with default assumptions concerning the gas collection system collection efficiency (75%) and an allowance for biological oxidation of methane in the soil cover (10%). This paper describes a numerical model developed to simulate heat, water, and gas, transport in landfill covers along with biological oxidation of methane in the upper layers of the soil cover profile. The model uses climatic conditions and models the change in soil gas permeability, the gas viscosity, and methane oxidation and other soil properties during the life of landfill cover. The model was used to simulate gas transport and methane oxidation in an example landfill cell. The bottom flux, from the waste mass into the soil cover, was simulated to be equivalent to 25% of the generated gas calculated using LANDGEM. Simulations were performed for 19 years using average daily climatic conditions in four climates.

Modeled surface emissions show a seasonal fluctuation due to the dependence of methane oxidation on climatic conditions. For the two cold climates (Iowa and Montana), surface emissions during winters are higher than that predicted by the default AP-42 method. Little to no biological activities occurs during the winters in these two climates. Biological oxidation, seems to occur even during winters in Florida and California climates. The modeled total emissions, total emissions during the 19-year period, in Florida were only 5.2% of those obtained using the AP-42 method. For the California, Iowa, and Montana, total emissions were 10.6%, 51%, 53% (respectively) of those obtained using the AP-42 method. The authors acknowledge that the model tends to over-estimate methane oxidation especially when the methane flux is ?very low, however, these results are an indication that the AP-42 guidelines might have to be revisited to reflect the effects of climatic conditions, soil types, and cover design on methane emission from landfills. Further work is being performed to better calibrate the model described in this paper using field observations.

INTRODUCTION

One way to estimate uncontrolled emissions of various compounds present in landfill gas is to use the AP-42 protocol proposed by the United States Environmental Protection Agency (USEPA, 1997) The widely used equation 5 in AP-42, when applied to landfill methane surface emissions, can be written as follows:

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$$CM_P = \left[UM_P * \left(1 - \frac{\eta_{col}}{100} \right) \right] * (1 - f_{ox}) \tag{1}$$

Where CM_P is the controlled mass emissions of methane (kg/year), UM_P is the methane generation from the waste mass in kg/year (estimated by LANDGEM or any other allowed model), η_{col} is the collection efficiency of the landfill gas collection system. The USEPA has allowed the use of 75% as the default collection efficiency for landfill. The term f_{ox} is the fraction of methane biologically oxidized by the cover soil. The USEPA has also allowed the use of 10% correction to account for methane oxidation that can occur in landfill cover soils. The use of 10% oxidation has not been well explained.

Methane oxidation rate has been reported to be a function of climate and soil type with higher oxidation rate associated with warmer climate and higher organic content. A numerical model, developed at Florida State University which combines water balance, heat transport models with a gas transport and oxidation component to estimate methane emission and oxidation from different landfill covers under different climatic conditions. One application of this model is to be used in conjunction of AP-42 guidelines but with a modeled methane oxidation instead of the 10% Default.

MODEL DESCRIPTION

In a landfill setting, water content, temperature, and barometric pressure are constantly changing depending on climate conditions, soil type, cover thickness, and vegetation. In order to better predict gas emissions and methane oxidation in landfill covers, the changing water content and temperature inside a cover has to be accounted for in any numerical or analytical solution. Numerical methods to estimate methane emission and oxidation have been limited to simulating column or bench-scale experiments where the soil water content and temperature are practically constant. A numerical model was developed by a research team at Florida State University that combines water and heat flow with a gas transport and oxidation model. The gas transport and oxidation model is able to use dynamic parameters associated with water content and temperature and incorporates dynamic methanotrophic activity. Surface emissions and oxidation can be estimated knowing cover design, management practices, and daily climatic conditions.

Daily volumetric water content and temperature are generated at several depths in the cover design by the unsaturated flow module. The gas transport module uses these dynamic results to simulate methane emission and oxidation during each day for the average climatic conditions along with the methane oxidation capacity of the cover soil.

Unsaturated Flow Modeling

The investigation of the volumetric water content and temperature profile of landfill cover on their performance involved simulations of water and heat flow in variably saturated soils using the computer program HYDRUS1D v3.0 (Simunek et al. 2005). The program numerically solves the Richards' equation for saturated–unsaturated water flow as follows:

$$\frac{\partial \theta}{\partial t} = \frac{\partial}{\partial x}[K(\frac{\partial h}{\partial x} + \cos\alpha)] - S \qquad (2)$$

where θ is volumetric water content, h is pressure head [L], x are the spatial coordinates [L], t is time [T], S is the sink term [$L^3 L^{-3} T^{-1}$], α is the angle between the flow direction and the vertical axis (i.e., $\alpha = 0$ for vertical flow, 90 for horizontal flow, and $0 < \alpha < 90$ for inclined flow), The hydraulic properties K [m s^{-1}] were represented by the Mualem-van Genuchten function (Van Genuchten, 1980).

$$S_e = [1 + (ah)^n]^{(1/n-1)} \qquad (3)$$

$$K_r = K_s S_e^l [1 - (1 - S_e^{1/(1-1/n)})^{(1-1/n)}]^2 \qquad (4)$$

where K_s is saturated hydraulic conductivity, K_r is relative hydraulic conductivity. l is a hydraulic conductivity parameter. a is related to the air-entry value [m^{-1}], and n is a pore size distribution parameter. *Se* in Eq. 3 is expressed by Eq. 5

$$S_e = \frac{\theta - \theta_r}{\theta_s - \theta_r} \qquad (5)$$

where θ_r is the residual water content [m^3 m^{-3}]. θ_s is the saturated water content [m^3 m^{-3}]

S in Eq. 2 is a sink term to account for water uptake by plant roots and is defined as:

$$S(h) = \alpha(h)T_p / L_R \qquad (6)$$

where, $\alpha(h)$ is the plant water stress function, T_p is the potential transpiration rate[LT^{-1}], L_R the depth [L] of the root zone.

Heat transport through the cover was coupled to water transport and is described with a convection-dispersion equation of the form:

$$\frac{\partial C_p(\theta)T}{\partial t} = \frac{\partial}{\partial x}[\lambda(\theta)\frac{\partial T}{\partial x}] - C_w\frac{\partial qT}{\partial x} - C_w ST \qquad (7)$$

where $\lambda(\theta)$ is the coefficient of the apparent thermal conductivity of the soil [M m s^{-3} K^{-1}]. $C_p(\theta)$ and *Cw* is the volumetric heat capacities [M m^{-1}s^{-2} K^{-1}] of the porous medium and the liquid phase, respectively. *q* is Darcian fluid flux density [m s^{-1}].

Gas Transport Modeling

Volumetric water content and temperature were generated at each node each day by HYDRUS1D. The gas transport model used these dynamic results to simulate methane emission and oxidation at each day. A continuity equation and a mass balance equation were used to describe the gas flow and reaction within the porous media.

$$\varepsilon \frac{dC_i}{dt} = -\frac{dJ_i}{dx} + r_i \qquad (8)$$

where ε is the air filled porosity [m^{-3}gas m^{-3}soil], C_i, the molar gas concentration [mol m^{-3}], J_i is the flux of gas component *i* including the diffusive and advective flux (mol m^{-2} s^{-1}), r_i is the reaction rate of gas component *i* [mol kg^{-1}dry soil s^{-1}], and *dt* [s] and *dx* [m], are time and vertical distance coordinates.

The flux of the gas, J_i, has two components: diffusion and advection. Gas diffusion in porous media is governed by Fick's law and gas advection is governed by Darcy's law, the total flux can then be expressed as follows:

$$J_i = -D_{soil,i} \frac{dC_i}{dx} - \frac{k}{\mu} \frac{dP}{dx} C_i \tag{9}$$

where the diffusion coefficient of gas component i, $D_{soil,i}$, in soil [m^2 s^{-1}]. k is the intrinsic permeability of the soil (m^2), μ is the gas mixture viscosity [N s m^{-2}] and P is the pressure [Pa]. The pressure P is obtained by the ideal gas law:

$$P = \sum_{i=1}^{4} C_i RT \tag{10}$$

where T is the absolute temperature (K) and R is universal gas constant [8.314 J K^{-1} mol^{-1}].

Methanotrophic Reaction

The reaction component of the gas transport equation was assumed to be the following (De Visscher and Van Cleemput 2003):

$$CH_4 + aO_2 \rightarrow bCO_2 \tag{11}$$

where a, b are the stoichiometric factors for oxygen and carbon dioxide, and were assumed to be 1.5 and 0.5. This leads to the following reaction equation:

$$CH_4 + 1.5O_2 \rightarrow 0.5CO_2 + 0.5CH_2O + 1.5H_2O \tag{12}$$
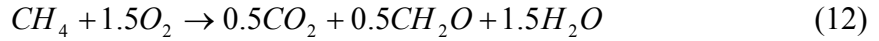
where CH_2O represents biomass. r_i in Eq. 7 is the reaction rate of methanotrophic bacteria, which is calculated by Michaelis-Menton kinetics from the incubation experiment.

$$r_{CH_4} = V_{max} \frac{C_{CH_4}}{K_{m[CH_4]} + C_{CH_4}} \frac{C_{O_2}}{K_{m[O_2]} + C_{O_2}} \tag{13}$$

where V_{max} is the maximum methane consumption rate [nmol s^{-1} kg^{-1}dry soil] and K_m is the half saturation constants [mol m^{-3}].

Dynamic Parameters

Air filled porosity ε, diffusion coefficient D, and gas permeability k are the function of volumetric water content as describe as follows:

$$\varepsilon(\theta) = \Phi - \theta \tag{14}$$

where Φ is the total porosity of soil. The diffusion coefficient of gas component i, $D_{soil,i}$, in soil can be calculated with the empirical model of Marshall (1959):

$$D_{soil,i} = r_{DC} D_{gas,i} = \frac{\varepsilon^{2.5}}{\Phi} D_{gas,i} \tag{15}$$

where r_{DC} is the relative diffusion coefficient, $D_{gas,i}$ is the diffusion coefficient of gas component i in gas-mixture and Φ is the total porosity (m^3void m^{-3}soil). Gas permeability change with water content can be described by Brooks and Corey (1964) equation that describes the dynamic gas permeability k as a function of the soil water saturation:

$$k = 0.0 \qquad \text{for } (u_a - u_w) \le (u_a - u_w)_b$$
$$k = k_0(1 - S_e)^2(1 - S_e^{(2+\lambda)/\lambda}) \quad \text{for } (u_a - u_w) > (u_a - u_w)_b \qquad (16)$$

where k_0 is the gas permeability for soil at a degree of saturation of zero (no water present), S_e is the effective degree of saturation defined by equation (16).

$$S_e = \frac{\theta - \theta_r}{\theta_s - \theta_r} \qquad (17)$$

where θ_r is the residual water content [$m^3\ m^{-3}$], θ_s is the saturated water content [$m^3\ m^{-3}$], u_a and u_w are the pore-air pressure and pore-water pressure (N m^{-2}), respectively, λ is the pore size distribution index, which is defined as the negative slope of the effective degree of saturation, S_e, versus matric suction, $(u_a - u_w)$ curve. The air entry value of the soil $(u_a - u_w)_b$ is the matric suction value that must be exceeded before air recedes in the soil pores.

A temperature correction was introduced for the biological oxidation parameters similar to De Visscher and Van Cleemput (2003). For the half saturation constant of methane, Km, the following equation was used (De Visscher and Van Cleemput, 2003):

$$f_{K,T} = \frac{0.00678 + 0.009814T}{\exp(1700(\frac{1}{T + 273.15} - \frac{1}{298.15}))} \qquad (18)$$

The maximum oxidation rate, V_{max} from one soil was measured at different temperatures (6°C, 15°C, 25°C, 33°C and 43°C). A temperature correction factor ($f_{V,T}$) was estimated for V_{max} as follow:

$$f_{V,T} = 2.235 - 0.18(T - 33) \ \text{(T>33)} \qquad (19)$$
$$f_{V,T} = 0.112T - 1.47 \ \text{(15<T<33)} \qquad (20)$$
$$f_{V,T} = 0.0142T \ \text{T<15} \qquad (21)$$

V_{max}, was also corrected for moisture content based on Boeckx et al. (1996). Their data shows a reduced oxidation rate when the moisture content is below 10% (weight based) due to physiological stress. Eqs 21 and 22 show the water content correction factor.

$$f_{V,m} = 10\frac{w}{\rho_{DB}} \ \text{(w<0.1)} \qquad (22)$$
$$f_{V,m} = 1 \ \text{(w>0.1)} \qquad (23)$$

where $f_{V,m}$ the moisture correction factor and w the moisture content ($g_{water}\ g_{drysoil}^{-1}$)

Boundary Conditions

Since the surface of the cover soil is open to the atmosphere, the gas composition above the surface node of the model was assumed to be the atmospheric gas compositions, which are 21.21% oxygen, 1.8 ppmv methane, 370 ppmv carbon dioxide and 78.75% nitrogen. However at the surface node of the model, the concentration will be different due to the mixing with the landfill gas (For a detailed explainaton see De Visscher and Van Cleemput (2003)):

$$C_{i,0} = C_{i,b} - \frac{J_{i,0}}{k_a} \tag{24}$$

where $C_{i,b}$ the background concentration of gas component i and $C_{i,0}$ the concentration of gas component i at the soil surface, k_a the mass transfer coefficient: 0.01m/s for moderate wind speeds.

The daily average barometric pressure is used by the model as a top pressure boundary condition. For the bottom boundary, a known pressure, or landfill gas flux (from the waste mass) can be used. In the modeling effort presented in this paper, a constant gas composition of the landfill gas flow coming into the cover due the flux boundary was assumed. The composition of the incoming flux into the bottom of the soil cover is assumed to be 60% CH_4 and 40% CO_2. This results in a no flow boundary for O_2 and N_2 and a fixed ratio between the CH_4 and CO_2 flux at the bottom. The temperature of the bottom boundary was assumed to stay constant (20 $^{\circ}C$).

APPLICATION OF MODEL WITHIN AP-42 GUIDELINES

To showcase how the presented model can be used to estimate methane surface emissions within AP-42 guidelines, an example landfill cell was assumed. The waste footprint of the example landfill cell was selected to be 35 acres. Filling history in the cell was obtained from a real landfill in the USA. Four climatic conditions were used for this study. Climatic data included daily precipitation, potential evapotranspiration (PET), maximum temperature, minimum temperature, and average temperature. Climate data were obtained for an average year in Florida, Iowa, Montana, and California. These climate data were used as a time-variable atmospheric boundary condition. The soil properties for HYDRUS1D were selected from HYDRUS1D default values for clayey loam. In order to minimize the influence of initial condition, a period of ten consecutive cycles of twelve months period was modeled by HYDRUS1D code. At the end of this period, daily water content and temperature in each node were obtained for the input of gas transport and oxidation model. The maximum soil oxidation capacity, V_{max}, was assumed to be 500 nmol kg^{-1} s^{-1}.

The USEPA LANDGEM was used as the default method to estimate methane generation rate in the assumed landfill cell. The collection efficiency of the gas collection system was assumed to be 75%. The remaining 25% of the generated methane was assumed to escape through the landfill cover being simulated in this study. The bottom boundary for the gas flow model was then set to equal 25% of the generated gas. Simulations were then performed to estimate surface emissions and methane oxidation from the example cell situated in all four climates.

Fig. 1 show the surface emissions predicted by the model described in this paper along with the emissions obtained using the default values within AP-42 guidelines (75% collection efficiency, 10% oxidation in the cover). The USEPA has allowed the use of 75% as the default collection efficiency for landfill. The USEPA has also allowed the use of 10% correction to account for methane oxidation that can occur in landfill cover soils.
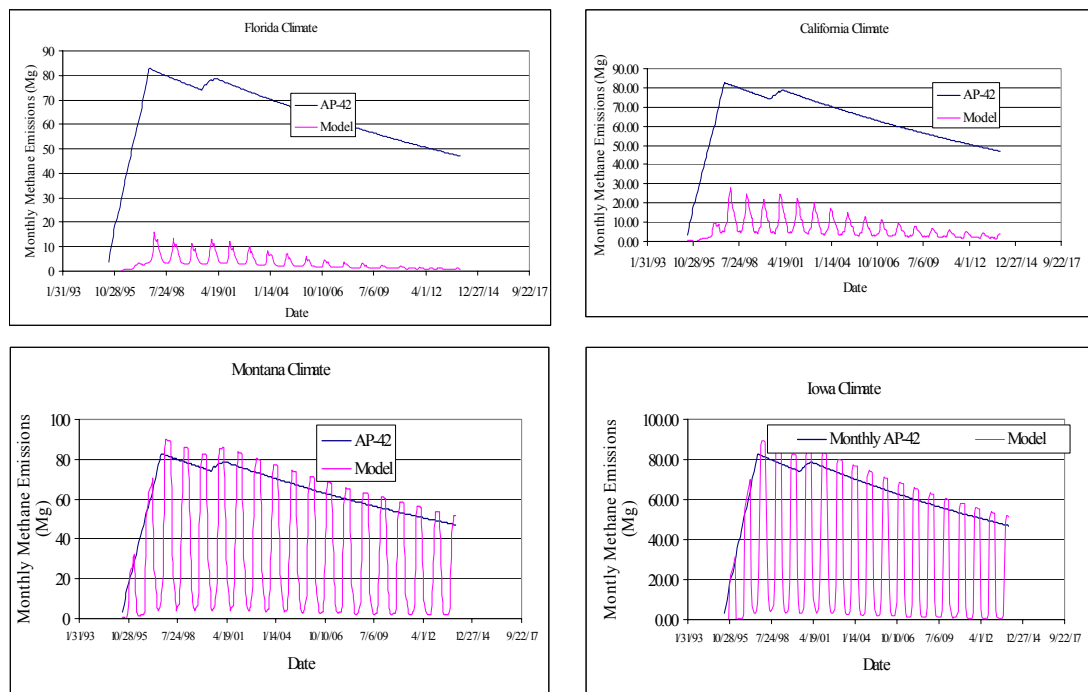
Fig. 1. Methane Emissions Predicted by Model and AP-42 Default Scenario for California, Florida, Montana, and Iowa.

Modeled surface emissions show a seasonal fluctuation due to the dependence of methane oxidation on climatic conditions. For the two cold climates (IA and MT) surface emissions during winters are higher than that predicted by the default AP-42 method. During the warm summers, however, the soil cover was able to keep surface emission close to zero. For the two warm climates (FL and CA), seasonal fluctuation in surface emissions are less dramatic since the soil cover stays relatively warm during the majority of the year. When considering the total amount of methane emitted during the entire simulated period (area under curve in Fig. 1), the modeled surface emissions were significantly lower than those obtained using the default AP-42 method. For instance, the modeled total emissions in Florida were only 5.2% of those obtained using the AP-42 method. For the California, Iowa, and Montana, total emissions were 10.6%, 51%, 53% (respectively) of those obtained using the AP-42 method. The authors acknowledge that the model tends to over-predict methane oxidation especially when the methane flux is ?very low, however, these results are an indication that the AP-42 guidelines might have to be revisited to reflect the effects of climatic conditions, soil types, and cover design on methane emission from landfills. Further work is being performed to better calibrate the model described here in using field observations.

CONCLUSIONS

A numerical model was described that allows the simulation of gas flow and methane oxidation under changing climatic conditions. The model predictions were compared to methane emissions obtained by AP-42 method using default assumptions on landfill gas collection system efficiency (75%) and the 10% methane oxidation in the soil cover. The model results show how the developed model reflects the change in methane oxidation in landfill covers with change in climatic conditions. Using, an example landfill cell, the modeled emissions were always significantly lower than those obtained using the AP-42 method. The modeled total emissions,

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total emissions during the 19-year period, in Florida were only 5.2% of those obtained using the AP-42 method. For the California, Iowa, and Montana, total emissions were 10.6%, 51%, 53% (respectively) of those obtained using the AP-42 method. Further research is being performed to calibrate the model using field observations.

ACKNOWLEDGEMENTS

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