# ESTIMATING METHANE EMISSION AND OXIDATION FROM EARTHEN LANDFILL COVERS

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**SUMMARY**: Measuring methane emission and oxidation through landfill covers has been done by many researchers. However, the numerical methods to estimate methane emission and oxidation are very limited. A simulation model was developed that combined water and heat flow model and a gas transport and oxidation model. The gas transport and oxidation model is able to use dynamic parameters associated with waster content and temperature and incorporates dynamic methanotrophic activity. Four sites were selected to showcase how emissions and oxidation can be estimated knowing cover design, management practices, and climatic conditions. Simulations were performed for scenarios with and without an active gas collection system. Different simulations were performed with and without organic amendments to the soil cover. Thirty-two simulations were conducted under different locations, climate condition, bottom pressure boundaries, and soil oxidation capacities.

Simulations showed that soil covers in subhumid areas can prevent high methane emission with blockage and decent oxidation capacity. In semiarid sites, higher emission was obtained due to the higher air filled void space of the soil. Oxidation capacities in semiarid sites are higher than those in subhumid sites since influxes of methane are higher in semiarid sites. High pressure underneath the cover caused higher emission in all sites. Even with active gas collection system (vacuum pressure), emissions were significant in semiarid climates. Soil oxidation is not only dependent on the potential methane oxidation capacity (Vmaxmax), but depends on methane availability.

## 1. INTRODUCTION

The global annual input of methane to the atmosphere is estimated to be 535±125 Tg (IPCC, 1995), of which about half is considered to be both anthropogenic and originating from biospheric processes, particularly anaerobic bacterial fermentation. Decomposition of refuse in municipal landfills is believed to be one of the major components of this biogenic methane, but past estimates of the emissions from this source have varied greatly, from 9 to 70 Tg per year. More reliable estimates are clearly needed, but it appears that landfills are the largest anthropogenic source of atmospheric methane in the United States and European countries. Rates for both methane production and oxidation can exceed observed rates for other terrestrial ecosystems by large factors. Field flux measurements (net emissions) vary over 7 orders of

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magnitude, from less than 0.0004 to about 4,000 grams per square meter per day (Bogner et al., 1997). These net emissions, of course, are the result of methane production, oxidation, and gaseous transport processes in the cover soil. Hence, it is difficult to predict emission rates at sites with various cover types, climatic regimes, and management practices. Oxidation rates in these soils range up to over 100 g $m^{-2}$ $d^{-1}$, and in some cases, the landfill can be a net sink for atmospheric methane oxidation. Emission data can be obtained by chamber, inert tracer, and micrometeorological methods.

Water content is a very important factor affecting $CH_4$ transport and oxidation in landfill cover soils. Boeckx et al. (1996) used a multiple linear regression analysis under different incubation conditions and concluded that water content has more influence on $CH_4$ oxidation than temperature. Christophersen et al. (2000) used statistical methods to analyze the effect of soil water content on $CH_4$ oxidation. They also concluded that water content can explain most of the variation observed in $CH_4$ emission data. Water plays three important roles. First, the optimum environment for $CH_4$ oxidizing bacteria (methanotrophy) is obtained at certain water content. Second, water content affects the penetration of oxygen ($O_2$) into the soils, which is the main reactor for $CH_4$ oxidation. As the water content increases in the soil, the $O_2$ diffusion into the soil is hindered. Thirdly, water content affects the air filled porosity of the soil and influences gas transport through the soil. As water fills the pores in the soil, it blocks the flow of gas upward. At the same time, the blocking of flow might lead to $CH_4$ emission due to the excess pressure built-up in the landfill (Boeckx et al., 1996).

Generally, $CH_4$ oxidation rate increases with increasing temperature (De Visscher et al., 2001). Low temperatures inhibit $CH_4$ oxidation (Whalen et al. 1990; Nozhevnikova et al., 1993; Borjesson and Svensson, 1997; Visvanathan et al., 1999). Borjesson and Svensson (1997) even reported that soil temperature is the controlling factor of $CH_4$ oxidation, and can explain 85% of the variation in measured $CH_4$ oxidation. Methanotrophic bacteria favor a certain range of temperatures. Czepiel et al. (1996) reported that oxidation rate increased as temperature increased to 36°C. They also reported that $CH_4$ oxidation essentially stopped when temperature reached 45°C. Humer and Lechner (2001) reported that $CH_4$ oxidation rate was 70-80% at 18°C. At a lower temperature of 4°C, little oxidation was observed. Borjesson and Svensson (1997) reported that the optimum temperatures for $CH_4$ oxidation were ~25 to 35°C.

This paper present a numerical model which combines water balance, heat transport models with a gas transport and oxidation model to estimate methane emission and oxidation from landfills with different climates. Four sites were selected to showcase how emissions and oxidation can be estimated knowing cover design, management practices, and climatic conditions. Simulations were performed for scenarios with and without an active gas collection system. Different simulations was performed with and without organic amendments to the soil cover.

## 2. MATERIALS AND METHODS

Four sites were selected to run the model for typical subhumid-warm weather (Florida), subhumid-cold weather (Iowa), semiarid-warm weather (California), and semiarid-cold weather (Montana). Soil cover properties are listed in 5.1. In the wet areas (Florida and Iowa), the covers were built thicker, 135 cm and 165 cm, respectively, to limit percolation , while in the dry areas, the covers were build thinner of 120cm thick each. All the covers are well vegetated.

### *2.1. HYDRUS1D Modeling Description*

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:

$$\frac{\partial \theta}{\partial t}=\frac{\partial}{\partial x}[K(\frac{\partial h}{\partial x}+\cos\alpha)]-S \qquad (1)$$

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 [$L^3L^{-3}T^{-1}$], $\alpha$ 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).

$$S_e=[1+(ah)^n]^{(1/n-1)} \qquad (2)$$

$$K_r=K_sS_e^l[1-(1-S_e^{1/(1-1/n)})^{(1-1/n)}]^2 \qquad (3)$$

where Ks is saturated hydraulic conductivity, Kr 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. 2 is expressed by Eq. 4

$$S_e=\frac{\theta-\theta_r}{\theta_s-\theta_r} \qquad (4)$$

where $\Theta_r$ is the residual water content [$m^3$ $m^{-3}$]. $\Theta_s$ is the saturated water content [$m^3$ $m^{-3}$]
S in Eq. 1 is a sink term to account for water uptake by plant roots and is defined as:

$$S(h)=\alpha(h)T_p/L_R \qquad (5)$$

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:

$$\frac{\partial C_p(\theta)T}{\partial t}=\frac{\partial}{\partial x}[\lambda(\theta)\frac{\partial T}{\partial x}]-C_w\frac{\partial qT}{\partial x}-C_wST \qquad (6)$$

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 $s^{-1}$]

### *2.2. 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.

$$\varepsilon\frac{dC_i}{dt}=-\frac{dJ_i}{dx}+r_i \qquad (7)$$

where $\varepsilon$ 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:

$$J_i = -D_{soil,i}\frac{dC_i}{dx} - \frac{k}{\mu}\frac{dP}{dx}C_i \qquad (8)$$

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$^2$), $\mu$ 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:

$$P = \sum_{i=1}^{4} C_i RT \qquad (9)$$

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

### *2.3. Methanotrophic Raction*

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

$$CH_4 + aO_2 \rightarrow bCO_2 \qquad (10)$$

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:

$$CH_4 + 1.5O_2 \rightarrow 0.5CO_2 + 0.5CH_2O + 1.5H_2O \quad (11)$$

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.

$$r_{CH_4} = V_{\max}\frac{C_{CH_4}}{K_{m[CH_4]} + C_{CH_4}}\frac{C_{O_2}}{K_{m[O_2]} + C_{O_2}} \qquad (12)$$

where $V_{\max}$ is the maximum methane consumption rate [nmol s$^{-1}$ kg$^{-1}{}_{\text{dry soil}}$] and $K_m$ is the half saturation constants [mol m$^{-3}$]. The Combined model simulation scheme is shown a flow chart (Fig. 1).

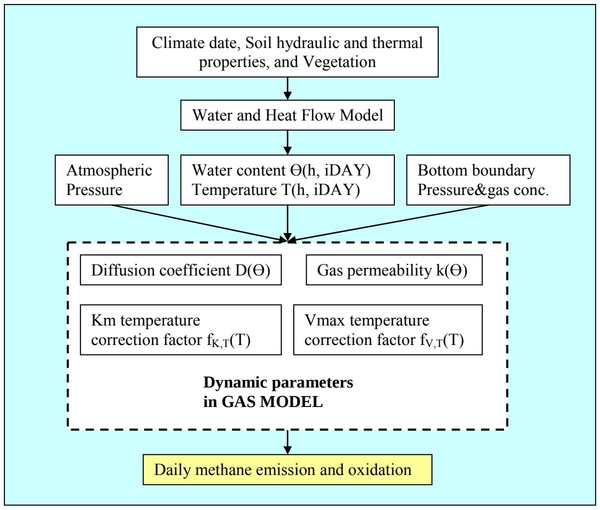

**Fig. 1. Flow chart of model algorithm.**

### *2.4. Boundary and Initial Conditions*

The gas composition at 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. For the bottom boundary, different constant pressures were used to describe the different situation of landfill pressure. Constant gas concentration boundary condition (60% methane and 40% carbon dioxide) was selected. The first day's initial condition was set up as all the node concentration equal to atmospheric gas concentration. The consequent initial condition at the beginning of each day was set equal to the pervious day's final concentration conditions.

### *2.5. Model Inputs and Assumptions*

Climatic input data for FL site was obtained from a weatherstation build nearby the site. Other sites' historical data were obtained from regional climate centers. The data included daily precipitation, potential evapotranspiration (PET), Maximum temperature, minimum temperature, and average temperature. These climate data were used as a time-variable atmospheric boundary condition. The soil properties for HYDRUS1D was measured for IA, MT, and CA sites (Table 1). The soil properties for the FL site were estimated based on soil classification. In order to minimize the influence of initial condition, a period of ten consecutive cycles of fourteen 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.

Thirty-two gas transport and oxidation simulations were conducted for different bottom pressure boundary condition and different soil oxidation capacity. Four kinds of pressure boundaries were used to represent different landfill conditions. (1) *High* pressure underneath the cover equal to constant 1.1 atm; (2) *Medium* pressure underneath the cover equal to constant 1.04 atm; (3) *Zero* pressure at bottom boundary equal to atmospheric pressure; (4) *Vacuum* pressure at the bottom boundary equal to -10 inches of water (2.49 kPa) as a typical vacuum pressure underneath cover when active landfill gas collection system is on. Two types of soil oxidation capacity were selected as: (1) High oxidation capacities as Vmaxmax=2000 nmol $kg^{-1}$ $s^{-1}$; (2) low oxidation capacity as Vmaxmax=500 nmol $kg^{-1}$ $s^{-1}$.

**Table 1. Landfill cover soil hydraulic properties summary.**

| **Site** | **Layer (top to bottom)** | **Thickness (cm)** | **$\alpha$ (1/cm)** | **n** | **$\theta r$** | **$\theta s$** | **Ks (cm/s)** |
|---|---|---|---|---|---|---|---|
| IA | Top soil | 60 | 0.0029 | 1.464 | 0.057 | 0.483 | $1.2\times10^{-5}$ |
| | Interim Cover | 105 | 0.0091 | 1.202 | 0 | 0.512 | $2.3\times10^{-5}$ |
| FL | Top soil | 75 | 0.036 | 1.56 | 0.078 | 0.54 | $2.33\times10^{-5}$ |
| | Clay cover | 60 | 0.036 | 1.56 | 0.068 | 0.45 | $1.19\times10^{-5}$ |
| CA | Surface layer | 60 | 0.028 | 1.31 | 0 | 0.3 | $2.9\times10^{-5}$ |
| | Storage layer | 45 | 0.028 | 1.31 | 0 | 0.3 | $2.9\times10^{-5}$ |
| | Interim layer | 15 | 0.028 | 1.31 | 0 | 0.3 | $2.9\times10^{-5}$ |
| MT | Surface layer | 15 | 0.0676 | 1.42 | 0 | 0.37 | $3.4\times10^{-5}$ |
| | Sandy silt | 45 | 0.0195 | 1.28 | 0 | 0.30 | $4.0\times10^{-5}$ |
| | Silty sand | 60 | 0.0711 | 1.45 | 0 | 0.36 | $7.9\times10^{-5}$ |

## 3. RESULTS AND DISCUSSION

### *3.1. Water Balance*

Water balance results are shown in Table 2. Fig. 2 also shows the typical water balance quantities (CA site). The total water applied to the cover (precipitation) was divided into four parts: runoff, evaporation, transpiration, and percolation. For semiarid areas (CA and MT), the cumulative precipitation for the entire period was 503 and 357.9 cm, respectively. In these semiarid areas, the runoffs were zeros since the precipitation was low and no heavy density of rainfall event exist. The transpiration of MT site was only 140.5 cm compared to 217.9 of CA site since the vegetation grow season of MT is shorter than CA. Overall percolation ratio over precipitation for CA and MT were 5.78% and 4.36%.

For the subhumid areas (FL and IA), the cumulative precipitation for the entire period was 1717.9cm and 977.1cm, respectively. The runoff of FL site was much higher than IA site since the rainfall was basically very dense in several events in FL. Transpiration and evaporation in FL was also higher than IA site, therefore the percentage of percolation over total precipitation for FL was 8.67%, which was lower than the IA site for 16.13%.

**Table 2. HYDRUS1D modeling results.**

| **Water balance (cm)** | **CA** | **FL** | **IA** | **MT** |
|---|---|---|---|---|
| Precipitation | 503 | 1717.9 | 977.1 | 357.9 |
| Runoff | 0 | 445.9 | 62.4 | 0 |
| Evaporation | 255.9 | 462.9 | 226.3 | 211.6 |
| Transpiration | 217.9 | 659.8 | 509.6 | 140.5 |
| Percolation | 29.1 | 149.3 | 157.7 | 15.6 |
| Percentage of Percolation | 5.78% | 8.67% | 16.13% | 4.36% |

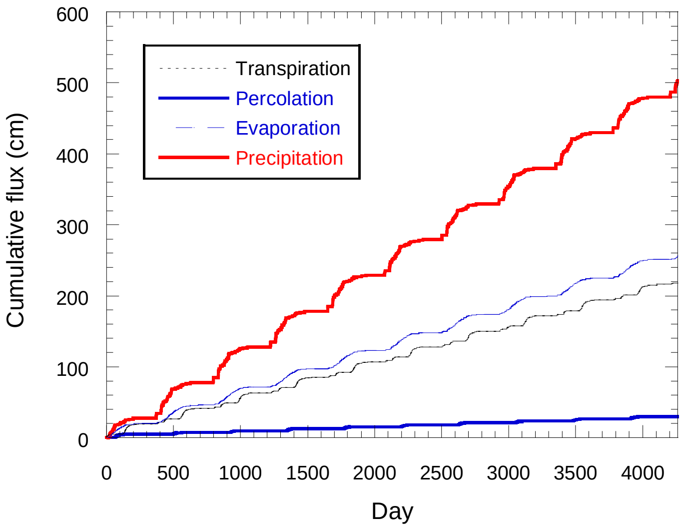

**Fig. 2. HYDRUS1D simulation of water balance (CA).**

### 3.2. Methane Emission and Oxidation Modeling

Fig. 3 shows 32 simulations with different pressure boundary condition and different Vmaxmax. Complete values are listed in Table 3. For semiarid sites (CA and MT), the overall emissions are higher than the wet sites (FL and IA), since there was more air filled void space for gas to migrate through the cover. Methane emission in MT site ranged from 2877.1~44904.1 g m-2 yr-1 and from 466~24152.6 g m-2 yr-1 in CA. However for the wet sites, the emission in FL was only 160.6~857.4 g m-2 yr-1 and 48.2~118 g m-2 yr-1 in IA. Lowest emissions were obtained when the bottom boundary is vacuum and with higher Vmaxmax for all sites. Highest emissions were obtained when the bottom boundary is high pressure (1.1 atm) and with low Vmaxmax.

For each site, high Vmaxmax leads to low emission and low Vmaxmax leads to high emission. The differences of methane emission due to Vmaxmax of Dry sites are larger than wet sites due to the higher influx of dry site. At medium pressure, the differences of emissions between high Vmaxmax and low Vmaxmax were 4.3, 163.5, 3520.1, and 5617.6 corresponding to influx values of 199.9, 3806.3, 21209, and 32744 for IA, FL, CA, and MT, respectively. The reason for phenomena is that low influx in wet sites had no enough methane for the methanotrophic bacterial to reach its oxidation capacity. So choosing a high oxidation capacity soil can be an efficient way to mitigate methane emissions, however for low emission site such IA, high oxidation capacity soil didn't significant reduced the emission.

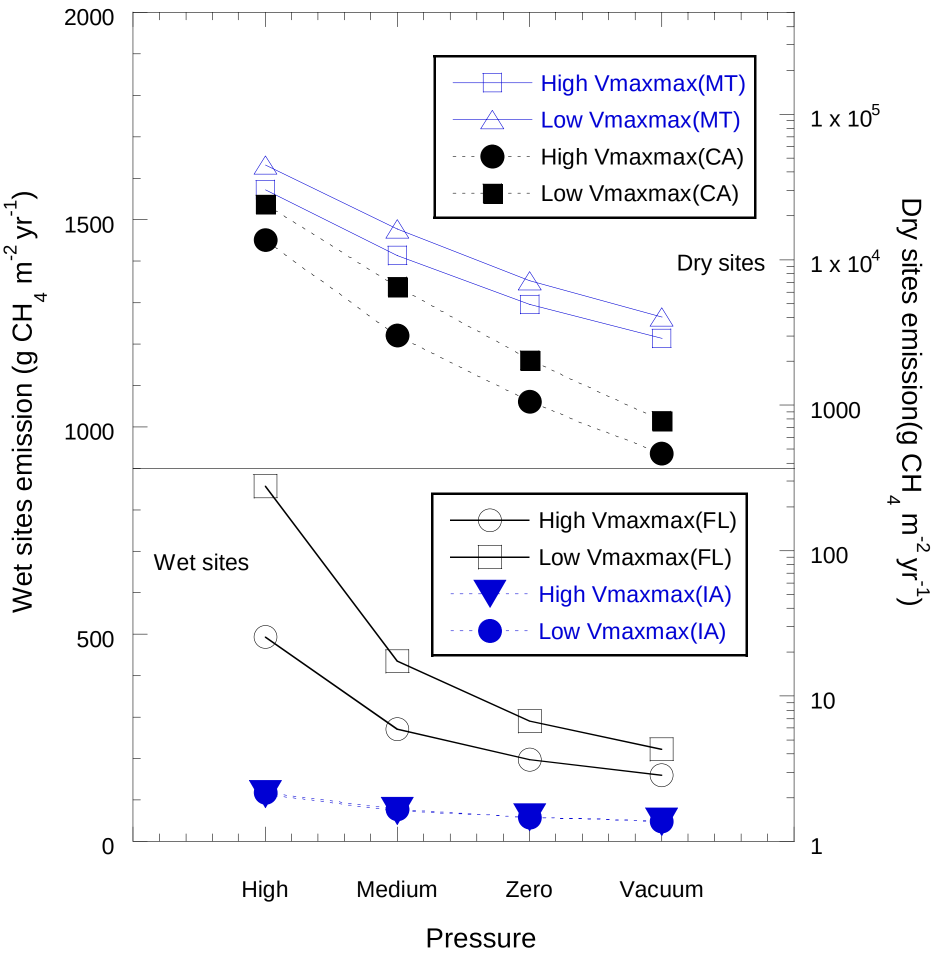

**Fig. 3 Yearly cumulative emission simulations summary for all sites.**

For each site, methane mass removed by oxidation increase with increasing of pressure. In FL site at low Vmaxmax, the mass removed is 2683.6, 2935.3, 3365.1, and 4098.1 g $CH_4$ $m^{-2}$ $yr^{-1}$ for vacuum, zero, medium, and high pressure, respectively.

Emissions from all sites increase with pressure increases. For FL site, the emissions of high pressure, medium pressure, zero pressure, and vacuum pressure were 857, 434.4, 290.9, and 221.9 g $m^{-2}$ $yr^{-1}$, respectively (low Vmaxmax). This show even with active landfill gas collection system, there was still emission of methane into the atmosphere due to the diffusion of methane. In dry sites, this emissions can be as high as 785.5 and 4039 g $m^{-2}$ $yr^{-1}$ for CA and MT site, respectively (low Vmaxmax).

**Table 3 summary of the yearly cumulative emission of all ET sites (g $m^{-2}$)**

| | **Pressure** | | IA | FL | CA | MT |
|---|---|---|---|---|---|---|
| **High Vmaxmax** | **High** | **Influx** | 264.5 | 5173.4 | 42795.6 | 62726.3 |
| | | **outflux** | 114.6 | 493.2 | 13724 | 30339 |
| | | **mass removed** | 149.9 | 4680.2 | 29071.6 | 32387.3 |
| | | **oxidation** | 56.67% | 90.47% | 67.93% | 51.63% |
| | **Medium** | **Influx** | 199.9 | 3806.3 | 21208.6 | 32744.4 |
| | | **outflux** | 73.6 | 270.9 | 3014.5 | 10657 |
| | | **mass removed** | 126.3 | 3535.4 | 18194.1 | 22087.4 |
| | | **oxidation** | 63.18% | 92.88% | 85.79% | 67.45% |
| | **Zero** | **Influx** | 168.2 | 3230.6 | 15254.6 | 20954 |
| | | **outflux** | 58.1 | 198 | 1056.8 | 4943.4 |
| | | **mass removed** | 110.1 | 3032.6 | 14197.8 | 16010.6 |
| | | **oxidation** | 65.46% | 93.87% | 93.07% | 76.41% |
| | **Vacuum** | **Influx** | 151.2 | 2909.4 | 10307.3 | 16412.2 |
| | | **outflux** | 48.2 | 160.6 | 466 | 2877.1 |
| | | **mass removed** | 103 | 2748.8 | 9841.3 | 13535.1 |
| | | **oxidation** | 68.12% | 94.48% | 95.48% | 82.47% |
| **Low Vmaxmax** | **High** | **Influx** | 264 | 4955.5 | 42359 | 63566.5 |
| | | **outflux** | 118 | 857.4 | 24152.6 | 44904.1 |
| | | **mass removed** | 146 | 4098.1 | 18206.4 | 18662.4 |
| | | **oxidation** | 55.30% | 82.70% | 42.98% | 29.36% |
| | **Medium** | **Influx** | 199.6 | 3799.5 | 21049 | 32277.8 |
| | | **outflux** | 77.9 | 434.4 | 6534.6 | 16274.6 |
| | | **mass removed** | 121.7 | 3365.1 | 14514.4 | 16003.2 |
| | | **oxidation** | 60.97% | 88.57% | 68.96% | 49.58% |
| | **Zero** | **Influx** | 168 | 3226.2 | 15199.2 | 20759.5 |
| | | **outflux** | 59.1 | 290.9 | 2030.8 | 7195 |
| | | **mass removed** | 108.9 | 2935.3 | 13168.4 | 13564.5 |
| | | **oxidation** | 64.82% | 90.98% | 86.64% | 65.34% |
| | **Vacuum** | **Influx** | 151 | 2905.5 | 10286 | 16300.4 |
| | | **outflux** | 48.9 | 221.9 | 785.5 | 4039 |
| | | **mass removed** | 102.1 | 2683.6 | 9500.5 | 12261.4 |
| | | **oxidation** | 67.62% | 92.36% | 92.36% | 75.22% |

## 4. CONCLUSIONS

A model was developed to study how landfill final earthen cover's construction and climate conditions affect methane emission and oxidation from landfills. This numerical model combines the water and heat flow model (HYDRUS1D) and a gas transport and oxidation scheme. The gas transport and oxidation model is able to incorporate dynamic parameters associated with water content and temperature, such as, air filled porosity, diffusion coefficient, and gas permeability. A dynamic oxidation capacity parameter (Vmax) is also embedded in the gas transport scheme. Four sites were selected to showcase how emissions and oxidation can be estimated knowing cover design, management practices, and climatic conditions. Simulations were performed for scenarios with and without an active gas collection system. Different simulations was performed with and without organic amendments to the soil cover

Simulations showed that soil covers in subhumid areas can prevent high methane emission with blockage and decent oxidation capacity. In semiarid sites, higher emission was obtained due to the higher air filled void space of the soil. Oxidation capacities in semiarid sites are higher than those in subhumid sites since influxes of methane are higher in semiarid sites. High pressure underneath the cover caused higher emission in all sites. Even with active gas collection system (vacuum pressure), emissions were significant in semiarid climates. Soil oxidation is not only dependent on the potential methane oxidation capacity (Vmaxmax), but depends on methane availability. The simulations are however the results of a numerical analysis. A verification of the results of the predictions of the developed model should be performed.

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