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# Numerical modelling of methane oxidation efficiency and coupled water-gas-heat reactive transfer in a sloping landfill cover

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## ABSTRACT

Microbial aerobic methane oxidation in unsaturated landfill cover involves coupled water, gas and heat reactive transfer. The coupled process is complex and its influence on methane oxidation efficiency is not clear, especially in steep covers where spatial variations of water, gas and heat are significant. In this study, two-dimensional finite element numerical simulations were carried out to evaluate the performance of unsaturated sloping cover. The numerical model was calibrated using a set of flume model test data, and was then subsequently used for parametric study. A new method that considers transient changes of methane concentration during the estimation of the methane oxidation efficiency was proposed and compared against existing methods. It was found that a steeper cover had a lower oxidation efficiency due to enhanced downslope water flow, during which desaturation of soil promoted gas transport and hence landfill gas emission. This effect was magnified as the cover angle and landfill gas generation rate at the bottom of the cover increased. Assuming the steady-state methane concentration in a cover would result in a non-conservative overestimation of oxidation efficiency, especially when a steep cover was subjected to rainfall infiltration. By considering the transient methane concentration, the newly-modified method can give a more accurate oxidation efficiency.

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## 1. Introduction

Methanotrophic microorganisms existed in landfill cover has been utilized to reduce the emission of methane (Scheutz et al., 2009; Czepiel et al., 1996). This biological reaction is considered to be a cost-effective method for mitigating methane emission, especially for small or/and old landfill, where installing a gas collection system may not be financially viable due to relative low methane gas generated. Microbial aerobic methane oxidation involves complex multi-physical processes in relation to coupled water-gas-heat transfer and microbial biochemical activities in unsaturated soil (Czepiel et al., 1996; Ng et al., 2015). Methane oxidation has been revealed to be significantly affected by the temperature and water content in soil (Abichou et al., 2011; Scheutz et al., 2009). Despite wide application of the use of microorganism to control methane emission, the mechanisms involved in the coupled bio-chemo-hydro-thermal processes are not clear. Although various numerical models that consider methane oxidation have been developed, most of them focused on the transfer of different

gases only and ignored the effects of water and heat transfer (Stein et al., 2001; Molins et al., 2008; De Visscher and Van Cleemput, 2003; Yuan et al., 2009). While there exists a limited number of models that incorporated water and heat transfer, the effects of water and temperature on the microbial activity were generally neglected (Garg and Achari, 2010; Hettiarachchi et al., 2007).

To simplify methane oxidation, one-dimensional (1-D) numerical process has been conducted (Garg and Achari, 2010; Hettiarachchi et al., 2007; Yuan et al., 2009; Spokas et al., 2011; Abichou et al., 2015). The 1-D assumption is valid only for the case of flat landfill covers, but it is not possible to study the two-dimensional (2-D) process involved in the sloping side of a landfill cover. This is because the 2-D spatial distribution of soil water content could potentially lead to variations of gas and heat transfer and methane oxidation rate, due to the coupled processes involved in the soil. The field measurements reported by Di Trapani et al. (2013) and Geck et al. (2016) show that methane emission in the upslope of a sloping cover is higher than that in the downslope. Unfortunately, any corresponding variations of soil water content and soil temperature with time were not measured. Hence, any interrelationship between the coupled water-gas-heat transfer and methane oxidation are not clear (Di Trapani et al., 2013; Geck et al., 2016; Garg and Achari, 2010; Hettiarachchi et al.,

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2007). More investigations are needed to reveal the underlying mechanisms involved in the microbial methane oxidation and its efficiency in a sloping cover, especially when different cover angles are considered (Di Trapani et al., 2013; Geck et al., 2016).

This study aims to investigate the methane oxidation efficiency in a sloping unsaturated landfill cover through 2-D numerical simulations. A fully coupled model, which can consider water-gas-heat reactive transfer during the biochemical reaction of methane oxidation in unsaturated soil (Ng et al., 2015), was adopted. The model was implemented in the finite-element based, multi-physics software COMSOL (COMSOL 5.2, 2015). The numerical model and input parameters were calibrated against a set of 2-D flume model tests that quantified methane oxidation (Berger et al., 2005; Berger, 2008) by comparing the computed results with measurements. Once calibrated, the same set of input parameters were used to carry out parametric study. This aims to identify critical factors that could affect the methane oxidation efficiency, including the angle of cover, rainfall intensity and landfill gas generation rate. A new method that considers transient changes of methane concentration during the estimation of the methane oxidation efficiency was proposed and compared against existing methods.

## 2. Methods

### 2.1. Theoretical model

The theoretical model includes the governing equations for 2-D transport of water, heat and gases of nitrogen ( $N_2$ ), oxygen ( $O_2$ ), carbon dioxide ( $CO_2$ ) and methane ( $CH_4$ ), by coupling the principles of mass conservation, energy conservation and fluid transport. Detailed model development is given in Ng et al. (2015).

#### 2.1.1. Water transfer

According to water mass balance, water transfer can be modeled using Richards' equation (Richards, 1931) considering water generation by methane oxidation:

$$\rho_w \frac{\partial \theta_w}{\partial t} = -\nabla(\rho_w v_w) + \rho_{DB} M_{H_2O} r_w \quad (1)$$

where  $\rho_w$  is water density;  $\theta_w$ ,  $\rho_{DB}$  and  $M_{H_2O}$  are volumetric water content, soil dry density and water molar mass, respectively;  $v_w$  is water flow velocity;  $r_w$  is water generation rate per unit mass of dry soil; and  $t$  is time. The physical meaning of Eq. (1) is that the soil water content change ( $\rho_w \frac{\partial \theta_w}{\partial t}$ ) is caused by the net water influx ( $-\nabla(\rho_w v_w)$ ) and water generation ( $\rho_{DB} M_{H_2O} r_w$ ) by methane oxidation.

Non-isothermal water flow ( $v_w$  in Eq. (1)) in unsaturated soil is described by Darcy's law as follows (Childs, 1969):

$$v_w = -k_w \left( \nabla \frac{P_w}{\rho_w g} + 1 \right) \quad (2)$$

where  $k_w$  is water permeability function;  $P_w$  is water pressure; and  $g$  is gravity acceleration.

#### 2.1.2. Multi-component gas transfer

Considering the principle of mass conservation for gas  $k$  (Molins and Mayer, 2007):

$$\frac{\partial}{\partial t} \left[ (1 - S_w) \phi c_g^k + S_w \phi c_w^k \right] = -\nabla[v_g c_g^k] - \nabla[v_w c_w^k] - \nabla N_g^k \pm \rho_{DB} r_g^k \quad (3)$$

where  $\phi$  and  $S_w$  is soil porosity and degree of saturation, respectively;  $c_g^k$  is molar concentration of gas  $k$ ;  $c_w^k$  is molar concentration of gas  $k$  dissolved in water;  $v_g$  and  $N_g^k$  are advective velocity of the gas mixture and the diffusive flux of gas  $k$  in the gaseous phase,

respectively;  $r_g^k$  is reaction rate per unit of dry soil mass for gas  $k$ . Eq. (3) considers that the transfer mechanisms of each gas component include (i) advection in the gaseous phase; (ii) advection of the dissolved gas  $k$  in water and (iii) gas diffusion in the gaseous phase. In Eq. (3),  $r_g^k$  is a function of soil temperature (details given later).  $S_w$ ,  $v_g$  and  $N_g^k$  are also affected by soil temperature through the thermal effects on soil water characteristic curve, permeability function and diffusion coefficient, respectively (Ng et al., 2015).

In Eq. (3), the molar concentration of gas  $k$  dissolved in water ( $c_w^k$ ) can be described by Henry's law (Reid et al., 1987)

$$c_w^k = H_g^k r_g^k \quad (4)$$

where  $H_g^k$  is Henry's coefficient (*dimensionless*) for gas  $k$ .

Ignoring gravitational effects, advective velocity for the gas mixture  $v_g$  in unsaturated soil can be described by Darcy's law (Parker, 1989):

$$v_g = -k_g \nabla \frac{P_g}{\rho_g g} \quad (5)$$

where  $P_g$  is gas pressure;  $k_g$  is gas permeability function; and  $\rho_g$  is gas density and can be determined by:

$$\rho_g = \sum_{k=1}^4 c_g^k M_g^k \quad (6)$$

where  $M_g^k$  is molar mass of gas  $k$ . In this study, the molar masses of  $O_2$ ,  $N_2$ ,  $CO_2$  and  $CH_4$  are considered to be  $3.2 \times 10^{-2}$  kg/mol,  $2.8 \times 10^{-2}$  kg/mol,  $4.4 \times 10^{-2}$  kg/mol and  $1.6 \times 10^{-2}$  kg/mol, respectively (Reid et al., 1987).

By Dalton's law and the ideal gas law (Reid et al., 1987),

$$P_g = \sum_{k=1}^4 c_g^k RT \quad (7)$$

where  $R$  is the ideal gas constant ( $8.314 \text{ J K}^{-1} \text{ mol}^{-1}$ ); and  $T$  is temperature.

The diffusive flux  $N_g^k$  can be described as follows (Bird et al., 1960):

$$N_g^k = -D_s^k \nabla c_g^k \quad (8)$$

where  $D_s^k$  is diffusion coefficient of gas  $k$  in the gas mixture through soil, which is mainly affected by gas concentration and soil water content (see Eqs. (S1)–(S3) in the supplementary document).

#### 2.1.3. Heat transfer

Using a similar approach adopted by Thomas and Ferguson (1999), invoking the principle of energy balance yields

$$\frac{\partial [E(T - T_r)]}{\partial t} = -\nabla(-\lambda_T \nabla T + Q_{conv}) + Q_{oxi} \quad (9)$$

where  $T_r$  is reference temperature;  $E$  is heat capacity of the soil at  $T_r$ ; and  $\lambda_T$  is thermal conductivity of soil. The heat transfer mechanisms considered in Eq. (9) include heat conduction ( $-\lambda_T \nabla T$ ), heat convection ( $Q_{conv}$ ) and heat generation by methane oxidation ( $Q_{oxi}$ ).

According to Thomas and Ferguson (1999), the heat capacity of soil at the reference temperature can be defined as follows:

$$E = (1 - \phi) \rho_s H_s + \phi S_w \rho_w H_w + \phi S_g \sum_{k=1}^4 M_g^k c_g^k H_{g,k} \quad (10)$$

where  $H_s$ ,  $H_w$  and  $H_{g,k}$  correspond to the specific heat capacities of soil particles, water and gas  $k$ , respectively; and  $\rho_s$  is soil particle density.

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