## ARTICLE IN PRESS

Geotextiles and Geomembranes xxx (2014) 1-9

Contents lists available at ScienceDirect



# Geotextiles and Geomembranes

journal homepage: www.elsevier.com/locate/geotexmem

# Organic acid transport through a partially saturated liner system beneath a landfill

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#### ARTICLE INFO

Article history: Received 14 April 2014 Received in revised form 9 June 2014 Accepted 21 June 2014 Available online xxx

Keywords: Landfill Composite liner Organic contaminant Millington and Quirk model Unsaturated diffusion model

#### ABSTRACT

A one-dimensional model was developed to investigate the transport of organic acids (commonly found in landfill leachate) through a partially saturated composite liner system beneath a landfill. Specific attention was paid to the influence of water content distribution on aqueous-phase diffusion process. Composite liner system was investigated, which was consisted of a geomembrane and a compacted clay liner underlain by three kinds of attenuation layer: sand layer, sandy clay loam layer, and clay layer. Volumetric water content profile in soil layers was obtained by Van Genuchten model, and the Millington and Quirk model was employed to describe the non-linear relationship between volumetric water content and diffusion coefficient. Three cases were analyzed and compared, i.e., totally saturated condition, unsaturated condition without considering unsaturated diffusion model, and unsaturated condition considering unsaturated diffusion barrier to organic contaminant due to its low water retention capacity. When the dependence of diffusion coefficient on volumetric water content is sufficiently considered, the contaminant flux decreases significantly in all the three kinds of attenuation layer. Unsaturated diffusion model capturing the relationship between water content and diffusion coefficient

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#### 1. Introduction

In modern landfills, liners are crucial horizontal barriers designed to protect groundwater and soils. Composite liners involving a geomembrane (GMB) over a compacted clay liner (CCL) or a GMB over a geosynthetic clay liner (GCL) are widely used in landfills, and extensive research has been conducted to investigate their short- and long-term performance (Rowe, 2005; Rowe, 2012). Intact HDPE GMBs without defects are excellent barriers to the advective and diffusive migration of many contaminant compounds in landfill leachate. Volatile organic contaminants (VOCs) can readily diffuse through HDPE GMBs (Haxo, 1990; Park and Nibras, 1993; Sangam and Rowe, 2001; Edil, 2003; Rowe et al., 2004; Sangam and Rowe, 2005; McWatters and Rowe, 2010), thus the penetration of organic contaminant could occur in the whole scale of composite liner. However, VOCs will volatilize in the

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http://dx.doi.org/10.1016/j.geotexmem.2014.06.007

unsaturated pore space and their transport will be different to other less volatile organic contaminants.

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Landfill leachate contains a variety of concentrated organics such as carboxylic acids (Kjeldsen et al., 2002). The transport process of these organic compounds through intact composite liners deserves careful consideration.

Although geomembrane wrinkles are commonly generated during construction, field observation suggested that wrinkles could be effectively controlled when geomembranes are covered at appropriate time and with sufficient restrained area (Chappel et al., 2012; Rowe et al., 2012). Overburden pressure caused by solid wastes mass as well as overlying drainage layers greatly contributes to the reduction of wrinkles (Dickinson and Brachman, 2006; Brachman and Gudina, 2008a,b). With good construction quality assurance, the majority of the geomembrane will be intact and in good direct contacted to subgrade and diffusion may be the dominated mechanism controlling the transport of organic contaminant beneath landfill. Therefore, the ability of geomembrane (GMB) to control the transport of organic contaminant requires quantification, and the capability of underlying soil layers to block contaminant diffusion needs careful consideration.

Please cite this article in press as: Liu, T., Hu, L., Organic acid transport through a partially saturated liner system beneath a landfill, Geotextiles and Geomembranes (2014), http://dx.doi.org/10.1016/j.geotexmem.2014.06.007

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Several analytical and numerical methods have been proposed to investigate the contaminant transport in composite liners (Foose, 2002, 2010; Foose et al., 2002; Rowe et al., 2004; Rowe and Booker, 2005; El-Zein and Rowe, 2008; Chen et al., 2009; El-Zein et al., 2012; Xie et al., 2013). A widely used assumption is that soils beneath GMB are totally saturated. This assumption leads to conservative prediction of contaminant transport in soils, and the results are applied in engineering design. However, field research results (Bonaparte and Gross, 1990; King et al., 1993) showed that a zone of partial saturation is a commonplace occurrence beneath a landfill. In western areas of China, groundwater level is low all the year. The unsaturated zone beneath the surface is guite thick and the soil layers are in low water content (Yang et al., 2012). Even for a saturated compacted clay liner, Celik et al. (2009) demonstrated that failure to consider the presence of an unsaturated zone beneath a clay liner could significantly underestimate the leakage through the liner. Normally, in landfill site selection and liner system design, a distance between landfill liner and groundwater level is required in order to minimize the contamination risk to groundwater. The potential effect of unsaturated zone on contaminant transport beneath landfills requires adequate evaluation. Typical analytical and numerical methods mainly focus on performance of composite liners, but fail to address the effectiveness of the composite liners in terms of ultimate impact on groundwater quality (Foose, 2010). Computational tools, such as POLLUTE (Rowe and Booker, 2005) and SPAS (El-Zein et al., 2012), are widely recognized as robust tools in dealing with the complexities and variations in landfill liner system. However, the potential effect of unsaturated condition for liner soils is not systematically investigated.

The effect of water content on aqueous-phase diffusion in soils was initially studied by soil scientists concerning nutrient supply, but it remained largely unexamined by geotechnical engineers in the context of contaminant transport and waste disposal (Hu and Wang, 2003). Several theoretical models were developed to quantitatively describe the diffusion behavior of solute in unsaturated soils (Lim et al., 1998; Revil and Jougnot, 2008). Although difficulties in matrix control are often encountered in laboratory tests on unsaturated soils, extensive laboratory studies concerning solute transport in unsaturated soils have been conducted (Rowe and Badv, 1996; Hutchison et al., 2003; Badv and Faridfard, 2005; Hamamoto et al., 2009, 2012; Savoye et al., 2010, 2012; Chou et al., 2012). Theoretical and experimental progress greatly helps to quantify and harness the slow diffusion process in unsaturated soils. A review of the unsaturated diffusion models describing relation between effective diffusion coefficient and volumetric water content was presented by Hu and Wang (2003). Chou et al. (2012) evaluated the solute diffusion models with tortuosity factor based on experiments on sand, sandy loam, and clay under varied degree of saturation, and concluded that the Millington and Quirk (1961) model fitted best with the experimental results. It is clear that the diffusion coefficient in aqueous phase would be significantly reduced as soils become unsaturated. Factors attributed to this reduction may include: (1) increase in diffusion path length; (2) increased viscosity of liquid next to the soil surface; (3) increased ionic interaction along small pores and water films; (4) interconnectivity between water filled pores and water films (Lim et al., 1998). The pure diffusion process is highly related to the variation of water content and diffusion coefficient, thus the dependence of unsaturated diffusion coefficient on water content needs to be sufficiently considered in the contaminant prediction.

A numerical model was developed to investigate the influence of water content variation and the unsaturated diffusion on the transport of organic contaminant with low volatility and high aqueous solubility beneath a landfill. Acetic acid, an organic compound with relatively high concentration in landfill leachate in China, was chosen as the typical organic compound (Kjeldsen et al., 2002; Yue et al., 2007). A typical composite liner system consisted of a geomembrane and a compacted clay liner underlain by an attenuation layer was investigated. Three kinds of attenuation soil layer: sand layer, sandy clay loam layer and clay layer were separately studied. The Millington and Quirk model was employed to describe the non-linear relationship between effective diffusion coefficient and volumetric water content. Three calculation conditions, i.e., totally saturated, unsaturated condition with or without considering unsaturated diffusion model, were investigated.

#### 2. Mathematical model

The governing geometry of the problem is shown in Fig. 1. A compacted clay liner and an attenuation layer are beneath the geomembrane. The groundwater level is at the base of the attenuation layer, and the flow is assumed to be in horizontal direction.  $L_{g}$ ,  $L_{s1}$ ,  $L_{s2}$  and h are thickness of geomembrane, compacted clay liner, attenuation layer, and aquifer, respectively. The origin of the coordinate system is at the interface between the geomembrane and the underlying compacted clay liner. The positive z-axis is assumed downward. The thickness of the compacted clay liner  $(L_{s1})$ is defined as 0.75 m, which is the minimum thickness prescribed in the technical code by Chinese government (Chinese Ministry of Construction, 2007). The thickness of the attenuation layer  $(L_{s2})$  is 3 m, so that the total distance between the top of liner and underlying receptor aquifer is 3.75 m, which is the minimum distance allowed under Ontario Ministry of the Environment (1998). The aguifer is assumed to be 1.0 m deep.

In engineering practice, soils at landfill site would vary in a wide range. In order to investigate the influence of soil characteristics on the transport and tempo-spacial distribution of contaminant, three kinds of underlying attenuation layer, i.e., sand layer, sandy clay loam layer, and clay layer, are separately investigated and are denoted as Scenarios 1, 2, and 3, respectively, in the later discussion.

Acetic acid was chosen to represent the organic compound in landfill leachate. The concentration of acetic acid (and other fatty acids or carboxylic acids) in leachate could be significant, especially in the leachate produced by MSW with high organic content and in the relatively early stage of leachate production (Kjeldsen et al., 2002). Yue et al. (2007) investigated the organic component in young landfill and presented that concentration of pentanoic acid, hexanoic acid and several other carboxylic acids could exceed 1000 mg/L, and the concentration of acetic acid could even reach 4286 mg/L. The volatility of acetic acid is relatively low. Under

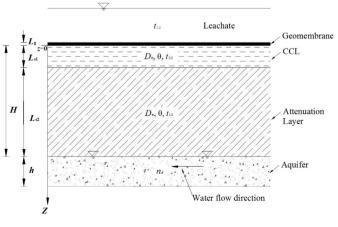


Fig. 1. Schematic diagram of diffusion through intact composite soil liner.

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