Contaminant transport through an unsaturated soil liner beneath a landfill

S.G. Fityus, D.W. Smith, and J.R. Booker

Abstract: Contaminant transport through soil is often modelled mathematically by means of the dispersion–advection equation. When investigating contaminant migration beneath a landfill, a common assumption is that the soil is saturated; however, it is well known that the soil beneath many landfills is only partially saturated. In this paper, steady-state unsaturated moisture distributions are employed in the dispersion–advection equation to find transient contaminant distributions beneath a landfill. A finite layer formulation is used to simplify the contaminant mass transport equation and account for heterogeneous soil profiles. Various assumptions concerning the flow regime beneath the landfill and the functional relation between volumetric water content and the diffusion coefficient in the transport equation are made to highlight differences between contaminant transport through saturated and unsaturated soils. It is found that diffusive contaminant mass transfer through a partially saturated soil liner is comparatively insensitive to variation of the volumetric moisture content at the top of the soil liner alone. However, accounting for the dependence of the diffusion coefficient on the volumetric moisture content of the soil does have a significant effect on diffusive mass transfer through a partially saturated soil liner. Though it is difficult to measure the dependence of the diffusion coefficient on the volumetric moisture content of the soil experimentally, this information appears necessary for a rational analysis of contaminant transport through partially saturated soils.

Key words: landfill, contaminant transport, unsaturated soil.

Résumé : Le transport de contaminants à travers le sol est souvent modélisé mathématiquement au moyen de l'équation de dispersion–advection. Au cours de l'étude de la migration de contaminants sous un enfouissement sanitaire, l'on pose habituellement l'hypothèse que le sol est saturé; cependant, il est bien connu que le sol sous plusieurs enfouissements sanitaires n'est que partiellement saturé. Dans cet article, des distributions non saturées de teneur en eau à l'état permanent sont utilisées dans l'équation de dispersion-advection pour déterminer les distributions transitoires de contaminants sous l'enfouissement sanitaire. Une formulation de couche finie est utilisée pour simplifier l'équation de transport de masse du contaminant et prendre en compte les profils de sol hétérogènes. L'on fait diverses hypothèses concernant le régime d'écoulement sous l'enfouissement et la relation fonctionnelle entre la teneur en eau volumétrique et le coefficient de diffusion dans l'équation de transport pour mettre en lumière les différences entre le transport de contaminant à travers les sols saturés et non saturés. L'on trouve que le transfert de masse des contaminants par diffusion à travers une membrane de sol partiellement saturé est comparativement insensible à la variation de la teneur en eau volumétrique au sommet de la membrane de sol seule. Cependant, le fait de prendre en compte la dépendance du coefficient de diffusion sur la teneur en eau volumétrique du sol a un effet significatif sur le transfert de masse par diffusion à travers une membrane de sol partiellement saturé. Quoique qu'il soit difficile de mesurer de façon expérimentale la dépendance du coefficient de diffusion sur la teneur en eau volumétrique du sol, cette information semble être nécessaire pour une analyse rationnelle du transport des contaminants à travers des sols partiellement saturés.

Mots clés : enfouissement sanitaire, transport de contaminants, sol non saturé.

[Traduit par la Rédaction]

Introduction

In order to protect the environment from contamination by pollutants, waste materials are now usually placed in engineered landfills. In recent years the design of engineered landfills has been regulated by the relevant authorities to ensure that the appropriate performance criteria are met. To demonstrate the likelihood of compliance with regulations, environmental engineers now make routine recourse to mathematical models of the proposed landfill. One outstanding feature of the modelling approach is the capacity for predictions to be made well into the future, thereby demonstrating the likely impacts of current practice on the environment and future generations. The validity of predic-

Received November 17, 1997. Accepted December 8, 1998.

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tions based on mathematical modelling of contaminant transport has been investigated and shown to be good for time periods measured in decades (Quigley and Rowe 1986) and even for time periods of thousands of years (Desaulniers et al. 1981; Quigley et al. 1983; Rowe and Sawicki 1992).

Moisture conditions beneath a landfill

There are many situations where the soil beneath a landfill is partially saturated. For example, many landfills are constructed in arid or semiarid environments, where the soil is usually partially saturated to great depths. The partially saturated conditions prevailing beneath landfills will be determined by a number of factors including the low-permeability capping layers and liners used to contain the waste.

In this paper, consideration is given to the two common situations of a natural soil liner and a geocomposite liner, where the geocomposite liner consists of a natural geologic material covered by a synthetic geomembrane. For municipal waste landfills, low- or high-density polyethylene (LDPE or HDPE) is often chosen as the geomembrane (Cossu 1994). Most geomembranes have a very low permeativity to water, with most moisture transfer through the membrane occurring by diffusion rather than by bulk flow (Artieres et al. 1994; Fityus and Smith 1998). The effect of a carefully laid geomembrane is to reduce moisture flow through the soil beneath the landfill to very small values (Giroud and Bonaparte 1989). If the piezometric groundwater table is located at some distance below the geomembrane and the air-entry value of the soil is not too high, then conditions are created where some depth of soil beneath the landfill will be in a state of partial saturation. Bonaparte and Gross (1990) investigated 55 landfills and reported that 53 had water tables below the base of the landfill. For this reason, it is expected that a zone of partial saturation beneath landfills is a commonplace occurrence. A documented example is the Keele Valley landfill in Ontario, Canada (King et al. 1993).

Based on previous engineering experience of impervious covers placed over partially saturated soils (e.g., Lytton 1969), it is expected that the moisture conditions in the soil beneath most of the landfill will ultimately attain a steady state. An important question arises as to the time period over which transient moisture change occurs and its influence on contaminant transport through the liner. To answer these questions comprehensively is beyond the scope of this paper. However, it is clear that very deep and initially very dry fine grained soils may take many decades to reach equilibrium moisture conditions beneath a landfill.

For the example landfill examined in this paper, which has an unsaturated zone 5 m in depth and employs parameters typical of a fine-grained soil, it can be demonstrated that the transient moisture changes occur over a time scale that is small relative to the time scale for contaminant mass transfer from the landfill. Therefore, from the viewpoint of an engineering contaminant transport design analysis, it is reasonable to assume that steady-state, unsaturated moisture conditions will prevail beneath the landfill, with only a small error being introduced by assuming that contaminant transport takes place through soil with steady-state moisture conditions.

Contaminant transport through unsaturated soils

Van Genuchten (1991) reviewed the extensive literature on solutions to transport equations for partially saturated soils, mainly published by soil and water-resources scientists, and found that the dispersion–advection equation is the most popular representation for contaminant transport, and Richards equation is most popular for the representation of moisture behavior. Few analytic solutions are known to the dispersion–advection equation in a partially saturated soil, but van Genuchten (1981) presented analytic solutions based on a spatially and temporally invariant unsaturated moisture content. Of course, for most applications of practical interest the moisture content of the soil varies with position.

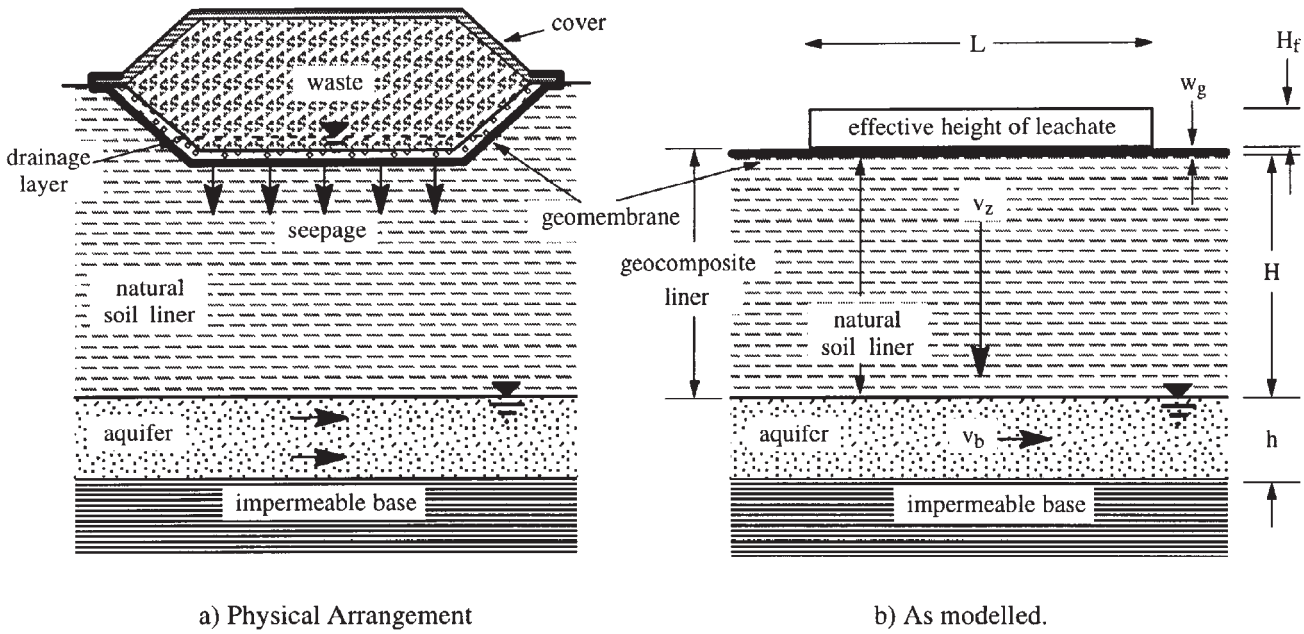
In this paper, a semianalytic approach to the solution of the dispersion–advection equation, specifically designed for the analysis of contaminant transport through horizontally layered soils beneath landfills, will be employed. This method has been developed by Rowe and others over a number of years (e.g., Rowe and Booker 1985*a*, 1985*b*, 1990; Rowe et al. 1995), and has proven robust and accurate for both one- and two-dimensional analysis of miscible contaminant transport through saturated soils. The solution technique employs a Laplace transformation to remove the time dependence from the governing equation, and a finite layer method to deal with heterogeneous soil profiles. Solutions are obtained in the real-time domain by numerical inversion of the transform solution (Talbot 1979).

The extension of contaminant transport analysis from saturated soils to unsaturated soils is relatively straightforward (e.g., Freeze and Cherry 1979). A simple but important change is the replacement of the soil porosity by the volumetric moisture content of the soil. In a partially saturated soil, this has the obvious effect of reducing the conductive cross-sectional area through which a contaminant may flow, but it is not immediately clear what effect this has on mass transfer through a soil liner beneath a landfill. The quantitative effect of partial saturation on mass transfer beneath the landfill is examined for the illustrative problem analyzed in this paper.

Contaminant transport analysis for a partially saturated soil is also influenced by the dependence of the diffusion, dispersion, and partitioning coefficients on the volumetric moisture content of the soil. Research relating diffusion coefficients for saturated soils (see review in Shackleford and Daniel 1991*a*, 1991*b*) to those for unsaturated soils was initially undertaken by soil scientists attempting to explain nutrient transfer to the roots of plants in the unsaturated zone (Klute and Letey 1958; Porter et al. 1960; Romkens and Bruce 1964; Warncke and Barber 1972; Barraclough and Tinker 1981). More recently, geotechnical engineers have investigated diffusion through unsaturated soils while attempting to explain the mass transfer characteristics of contaminants through leachate collection systems within landfills (Rowe and Badv 1996*a*, 1996*b*). The quantitative effect of the dependence of the diffusion coefficient on the volumetric moisture content of the soil is also examined in the illustrative problem analyzed in this paper.

It is also generally recognized that the dispersivity of a soil is a function of the volumetric moisture content of the soil. However, in the case of contaminant transport through a landfill liner functioning as intended by the design engi-

Fig. 1. Typical landfill arrangement.



neer, the advection is so small that the dispersivity may be taken as zero (Rowe et al. 1995), and this approximation will be adopted here.

The partitioning coefficient is a parameter in the dispersion–advection equation representing a geochemical model consisting of a single chemical equilibrium equation. Although it is well recognized that the partitioning coefficient can play a crucial role in controlling the rate of mass transfer through saturated soils and has been extensively investigated (Rowe et al. 1995), there is an unfortunate dearth of information on the effect of partial saturation on the partitioning coefficient. To the authors' knowledge, only two papers have tried to quantify the relationship between partial saturation and the partitioning coefficient for inorganic ions (Brown 1953; Lim et al. 1994). While this initial research has revealed that there is a significant dependence of the partitioning coefficient on the volumetric moisture content for at least some soils, the implications of this for geotechnical design of landfill liners are unclear. Experimental data quantifying the dependence of the contaminant partitioning coefficient on the volumetric moisture content of clay soils presents an important challenge for geotechnical experimentalists. For the illustrative example examined in this paper, it is assumed that there is no sorption onto the soil skeleton, although this may easily be incorporated in the analysis when appropriate data are available (Fityus and Smith 1997).

Additional considerations when analyzing contaminant migration through unsaturated soils

When a coarse-grained soil desaturates, the air phase initially forms as occluded air bubbles (Fredlund and Rahardjo 1993). As the degree of suction increases, the air phase in both fine- and coarse-grained soils becomes continuous and moisture movement by diffusive mass transfer along vapor partial pressure gradients becomes possible. The vapor diffusion coefficient depends on the moisture content of the soil, increasing as the moisture content decreases. However, it has been found that under isothermal conditions water vapor flow is small (Fredlund and Rahardjo 1993), so it is neglected in the analysis presented here.

Further, it is noted that, given suitable conditions, there are a number of subtle physical behaviors that can be exhibited by clay soils and are relevant to a contaminant transport analysis through unsaturated soils. One of these is osmotically induced fluid flow in response to contaminant concentration gradients. A montmorillonite soil, for example, can act as an imperfect semipermeable membrane. The effectiveness of a soil to act as a semipermeable membrane is measured by its osmotic efficiency parameter, with a value of one denoting perfect membrane efficiency and a value of zero denoting no osmotically induced fluid flow (Mitchell 1991). Experiments have also shown that the osmotic efficiency parameter of a clay is moisture dependent, increasing as the moisture content decreases. Kaolinitic clays, for example, have zero osmotic efficiency factor when saturated and for most of the range of partial saturation; however, kaolinitic clay begins to act as a imperfect semipermeable membrane at a volumetric moisture content of less than approximately 10% (Olsen 1972). In this paper, only soils that do not act as a semipermeable membrane are considered (i.e., the osmotic efficiency parameter is taken to be zero at all moisture contents).

Description of the example problem and analysis method employed in this paper

In this paper, consideration is given to the transport of a single, miscible, nonvolatile contaminant from a landfill, through a natural soil liner or a geocomposite liner under steady-state moisture conditions (Fig. 1). This leads to the consideration of a linear, second-order, partial differential transport equation with nonconstant coefficients in one spatial dimension. Perhaps the simplest approximate method for solving this equation is to divide the soil into many

sublayers and assume that for each sublayer the coefficients in the transport equation are constant, that is, assume that the volumetric moisture content is constant within each sublayer. In this paper, a more rigorous solution method is adopted. The nonconstant coefficient transport equation is solved directly in the Laplace transform domain using polynomials. Although this does not lead to a significant gain in computational efficiencies over the simpler sublayering method, it does provide a numerically independent method of solution, allowing greater confidence in the accuracy of the computed solutions.

It is expected that the contaminant mass transfer characteristics of a liner will be significantly influenced by the partial saturation of the soil. Landfill design will be improved by understanding and quantifying these effects. For the purposes of illustration, the performance of a landfill over a partially saturated natural soil is analyzed, and the following questions are addressed. (1) Given representative parameter values, what are the mass transfer characteristics of the landfill liner, assuming complete saturation of the soil? This solution is employed as a reference solution against which contaminant transport solutions for unsaturated soils are compared. (3) How does the moisture dependence of the diffusion coefficient influence the contaminant mass transfer through the soil liner? (4) What effects does partial saturation of the soil liner beneath a geomembrane have on contaminant mass transfer through a geocomposite liner? (5) How is the contaminant mass transfer through the partially saturated geocomposite liner affected by variations in the permeativity of the geomembrane? (6) What influence does a small advective flow through a partially saturated liner have on its mass transfer behavior?

Though it is believed that the trends revealed by the analysis of the illustrative problem presented here are general ones, the answers found to these questions naturally apply only to the specific example considered and should not be extrapolated to different design conditions. Every landfill design is unique and should be designed on an individual basis.

Moisture in an unsaturated soil liner

In the period following construction, the soil beneath a landfill may either wet up or dry out, depending upon the initial moisture condition of the soil and the hydraulic boundary conditions. The hydraulic flux through the top of the unsaturated soil liner can be estimated using a water-balance model of the landfill. The water-balance model should take into account a multitude of factors such as climate, material properties and geometry of the cap layer, surface grading, vegetation covering, waste type and initial moisture content, and waste and liner permeabilities, and the details of landfill operations such as leachate recycling and (or) landfill gas removal.

The lower hydraulic boundary condition for the unsaturated soil will be fixed by the position of the peizometric groundwater table and the air-entry value of the soil. The position of the groundwater table should in turn be estimated from a regional groundwater model, taking into account the regional hydrogeology and any likely changes in resource usage. It is noted that climate-change predictions could be included in these models.

It is also appreciated that biodegradation of the waste in a municipal landfill may lead to significant time-dependent temperature gradients, which in turn may lead to changes in the moisture profile of the surrounding soil. However, for the sake of simplicity, the position of the water table is taken to be time invariant and the temperature gradient zero in the illustrative example analyzed here. Given the estimated hydraulic boundary conditions for the unsaturated soil, it is necessary to estimate the moisture distribution through the partially saturated soil, and the period required for steady-state moisture conditions to become established.

There are many approaches to modelling moisture flow in unsaturated soils. Pullan (1990) presents a comprehensive overview of the substantial body of unsaturated flow theory which has been developed up to 1990. Fityus and Smith (1994) employed the so-called "*theta*–based" form of Richards equation to model unsaturated moisture flow, viz.,

$$[1] \qquad D_{\mathrm{m}}(\vartheta)\frac{\partial^2\theta}{\partial z^2} - \alpha D_{\mathrm{m}}(\vartheta)\frac{\partial\theta}{\partial z} = \frac{\partial\theta}{\partial t}$$

where z is the vertical position, t is time, ϑ is the volumetric moisture content of the soil, $D_{\mathrm{m}}(\vartheta)$ is the moisture diffusivity function, α is a parameter described by Philip (1968) as "a measure of the relative importance of gravity and capillarity for water movement (within the soil)," and θ is a new variable of mathematical convenience obtained from the application of a Kirchhoff transformation to the volumetric moisture content ϑ, viz.,

$$[2] \qquad \theta = \int_{\vartheta_*}^{\vartheta} D_{\mathrm{m}}(\vartheta)\mathrm{d}\vartheta$$

In eq. [2], ϑ_* is a convenient reference value, here taken to be dry soil (i.e., $\vartheta = 0$). Equation [1] may be employed to analyze the evolution of moisture profiles beneath the landfill given any initial condition. Steady-state moisture profiles may be estimated either as long-term solutions of eq. [1] or as a solution of the steady-state governing differential equation. In many cases, a good approximation to real soil behavior is found by adopting an exponential expression for the moisture diffusivity (Mualem 1976), viz.,

$$[3] \qquad D_{\mathrm{m}}(\vartheta) = D_{\mathrm{mo}}\mathrm{e}^{\gamma\vartheta}$$

where D_{mo} is the limiting diffusivity (as the moisture content approaches zero), and γ is an empirically fitted exponent. Substituting this approximation for the moisture diffusivity in eq. [2] results in a relationship between the true and Kirchoff–transformed moisture contents, viz.,

$$[4] \qquad \theta(\vartheta) = \frac{D_{\mathrm{mo}}(\mathrm{e}^{\gamma\vartheta} - 1)}{\gamma}$$

For a single layer, with moisture contents ϑ_{T} and ϑ_{B} at the top and bottom of the layer, respectively, the general steady-state solution to eq. [1] is

$$[5] \qquad \theta(z) = \frac{\theta_{\mathrm{B}} - \theta_{\mathrm{T}}\mathrm{e}^{\alpha H}}{1 - \mathrm{e}^{\alpha H}} + \left(\frac{\theta_{\mathrm{T}} - \theta_B}{1 - \mathrm{e}^{\alpha H}}\right)\mathrm{e}^{\alpha z}$$

where H is the thickness of the soil component in a geocomposite liner, and so the volumetric moisture distribution is here estimated to be given by

$$[6] \qquad \vartheta(z) = \frac{1}{\gamma}\ln\left\{\frac{\gamma\left[\frac{\theta_B - \theta_T e^{\alpha H}}{1 - e^{\alpha H}} + \left(\frac{\theta_T - \theta_B}{1 - e^{\alpha H}}\right)e^{\alpha z}\right]}{D_{mo}} + 1\right\}$$

and the Darcy flux, v_z, through the soil layer is given by

$$[7] \qquad v_z = a\,\frac{\theta_B - \theta_T e^{\alpha H}}{1 - e^{\alpha H}}$$

For a single layer, eqs. [6] and [7] can be evaluated directly. However, if the soil is horizontally stratified, continuity of moisture flux and free energy (i.e., soil suction) apply at the soil layer interfaces. Note that this implies the moisture profile may be discontinuous at the interfaces in a stratified soil.

Contaminant transport equation

The dispersion–advection equation is adopted as the governing differential equation describing contaminant transport through partially saturated soil. The form of the one-dimensional dispersion–advection equation with linear equilibrium controlled sorption is

$$[8] \qquad \vartheta D\frac{\partial^2 c}{\partial z^2} + \left(\vartheta\frac{\partial D}{\partial z} + D\frac{\partial \vartheta}{\partial z} - \vartheta v_t\right)\frac{\partial c}{\partial z} = \left(\vartheta + \rho_d K_d\right)\frac{\partial c}{\partial t}$$

where c is the concentration of contaminant in the pore fluid, v_t is the average true linear velocity of the pore fluid in the z direction, D is the effective hydrodynamic dispersion coefficient, ρ_d is the dry density of the soil, and K_d is the increment in mass of contaminant sorbed onto the soil skeleton per unit increment of concentration per unit dry mass of soil.

Equation [8] may be simplified by the introduction of the Laplace transformation defined by

$$[9] \qquad \bar{c}(s) = \int_0^\infty c(t)e^{-st}dt$$

where $\bar{c}(s)$ is the transformed concentration function in terms of the transform parameter s. Assuming the soil is initially contaminant free, eq. [8] is equivalent to

$$[10] \qquad \vartheta D\frac{\partial^2 \bar{c}}{\partial z^2} + \left(\vartheta\frac{\partial D}{\partial z} + D\frac{\partial \vartheta}{\partial z} - \vartheta v_t\right)\frac{\partial \bar{c}}{\partial z} = \left(\vartheta + \rho_d K_d\right)s\bar{c}$$

Of course, eq. [10] has to be solved subject to appropriate Laplace transformed boundary conditions. These conditions are discussed in detail as the need arises in the example problems.

An advantage of the proposed formulation is that eq. [10] is compatible with the integral transform techniques for solute transport analysis developed by Rowe et al. (1995). As will be shown, the solution is obtained in the Laplace transform domain using semianalytic methods and numerically inverted to find the solution in the real-time domain (Talbot 1979). For problems with two spatial dimensions, a Fourier transform may also be employed, thereby retaining the elegance of the analysis. Integral transform methods have the advantages of requiring minimal data preparation and of being computationally efficient, especially for evaluating solutions at large time periods and in two spatial dimensions. These advantages facilitate the rapid assessment of landfill design alternatives and so are useful as a design aid.

Finite layer formulation for contaminant transport analysis

Contaminant transport solution for a single soil layer

Equation [10] can be particularized to a given problem by choosing specific functions describing the spatial variation of ϑ and D. If spatially invariant values for the moisture content and diffusion coefficient are employed, then eq. [10] reduces to the familiar dispersion–advection equation for a saturated soil. This is computationally convenient, as contaminant transport through a saturated zone of soil (e.g., the capillary zone) is easily accommodated in the analysis.

Equation [6] is a relation describing the moisture content as a function of z, but it is of a difficult mathematical form and, if substituted directly in eq. [10], leaves this equation intractable. Although quadratic or higher degree polynomial approximations to the moisture distribution could be employed to approximate eq. [6], a simple alternative is to "sublayer" the soil, approximating the moisture distribution in each sublayer by a linear moisture distribution. Adopting this approach, the moisture distribution to be employed in the transport equation is

$$[11] \qquad \vartheta(z) = A + Bz$$

where A and B are empirically fitted constants. Note that the finite layer approach enables the soil moisture distribution to be broken into discrete segments, so regardless of the complexity of the actual moisture distribution in the soil liner, the simple linear expression of eq. [11] can always give a good approximation to the moisture distribution if small enough layers are used.

There is comparatively little research by geotechnical engineers on contaminant diffusion through soil under unsaturated conditions (Lim et al. 1994; Rowe and Badv 1996*a*, 1996*b*). More research has been conducted by soil scientists interested in nutrient movement in unsaturated soils. However, due to the experimental difficulties involved, only a handful of research papers exist on diffusion through partially saturated clay soils.

Based on the experimental data available (for example, see Porter et al. 1960; Rowell et al. 1967; Barraclough and Tinker 1981; Conca and Wright 1990), it is clear that the diffusion coefficient is significantly reduced as the soil becomes unsaturated, and this is assumed to reflect decreasing continuity and increasing tortuosity as the volumetric moisture content of the soil decreases. An examination of experimental data shows that a linear relation between the unsaturated diffusion coefficient and the volumetric moisture content is a reasonable approximation in many cases. In the numerical examples presented here, it is assumed that a bilinear model is adequate, and so

$$[12] \qquad D(\vartheta) = \begin{cases} \Lambda + \Omega\vartheta & \vartheta > \vartheta_1 \\ 0 & \vartheta \le \vartheta_1 \end{cases}$$

where $D(\vartheta)$ is the effective diffusion coefficient of the solute through the pore fluid, and Λ and Ω are empirically fitted

constants for a particular soil. Note that the constant Λ for soils may be negative, and so for the bilinear model the diffusion coefficient may be zero valued at a nonzero moisture content, ϑ_1. Based on experimental data, this appears to be a good approximation in many cases.

Now when eqs. [11] and [12] are employed in eq. [10], the transformed contaminant flux equation is

$$[13] \qquad \bar{f}(z) = -(Jz^2 + Lz + G)\frac{\mathrm{d}\bar{c}}{\mathrm{d}z} + E\bar{c} = 0$$

and substituting this into the mass conservation equation leads to the governing transport equation

$$[14] \qquad (Jz^2 + Lz + G)\frac{\mathrm{d}^2\bar{c}}{\mathrm{d}z^2} + (2Jz + L - E)\frac{\mathrm{d}\bar{c}}{\mathrm{d}z} - (X + Yz)\bar{c} = 0$$

where E, G, J, L, X, and Y are constants comprised of the parameters A, B, Λ, and Ω (and K_d if desired). Note that by setting the coefficient Ω to zero, eq. [14] can be used to examine unsaturated soil with a constant diffusion coefficient, and by setting B to zero implies an unsaturated soil with a spatially uniform moisture content. Of course, when Ω and B are zero and A is chosen to be the soil porosity, eq. [14] reduces to the usual transport equation for a saturated homogeneous soil.

Equation [14] is an ordinary, homogeneous differential equation with nonconstant coefficients. Solutions to this equation can be found using a power series, and so we let the Laplace-transformed contaminant concentration be approximated by

$$[15] \qquad \bar{c}(z) = \bar{a}_0 + \bar{a}_1 z + \bar{a}_2 z^2 + \ldots\ldots + \bar{a}_k z^k = \{\bar{a}_j\}^T\{z^j\}$$

where

$$[16] \qquad \{\bar{a}_j\}^T = \{\bar{a}_0, \bar{a}_1, \bar{a}_2, \ldots. \bar{a}_k\} \text{ and } \{z^j\}^T = \{1, z, z^2, \ldots.. z^k\}$$

It can be shown that coefficients of polynomials of degree 2 and higher can be expressed as a linear combination of the coefficients $\bar{a}_0$ and $\bar{a}_1$, that is,

$$[17] \qquad \{\bar{a}_j\} = \bar{a}_0\{\delta_j\} + \bar{a}_1\{\varsigma_j\}$$

where the vectors $\{\delta_\mathrm{j}\}$ and $\{\varsigma\}$ are found using matrix condensation after substitution of eq. [15] into eq. [14].

Then employing eq. [17] in eq. [15] shows

$$[18] \qquad \bar{c}(z) = \bar{a}_0\{\delta_j\}^T\{z^j\} + \bar{a}_1\{\varsigma_j\}^T\{z^j\}$$

where $\bar{a}_0$ and $\bar{a}_1$ are found from the boundary conditions. To improve the accuracy of the power-series approximation over the interval $0 \le z \le h$, a simple mapping of the interval $0 \le z \le h$ onto the interval $-1 \le z^* \le 1$ may be employed, viz.,

$$[19] \qquad z^* = \frac{(2z - h)}{(h)}$$

and the mapped concentration function c^* then approximated by a series of Chebyshev polynomials (T_k). The Chebyshev polynomials are $T_0 = 1$, $T_1 = z^*$, and, in general,

$$[20] \qquad T_{k+1}(z^*) = 2z^*T_k - T_{k-1} \qquad k \ge 1$$

The use of Chebyshev polynomials improves the accuracy of the series approximation by ensuring uniform convergence of the series over the approximation interval (Gerald and Wheatley 1984).

Global matrix for a multilayer soil profile

Now let the concentration at the top of the nth horizontal soil layer in an N-layered soil profile ($\bar{c}_{\mathrm{T}n}$) be expressed by eq. [18], and likewise the concentration at the bottom of the layer ($\bar{c}_{\mathrm{B}n}$). Similarly, the contaminant flux at the top ($\bar{f}_{\mathrm{T}n}$) and bottom ($\bar{f}_{\mathrm{B}n}$) of the soil layer may be expressed using eq. [13]. From this set of four equations the constants $\bar{a}_0$ and $\bar{a}_1$ may be eliminated, leading to

$$[21] \qquad \begin{Bmatrix} \bar{f}_{\mathrm{T}n} \\ \bar{f}_{\mathrm{B}n} \end{Bmatrix} = \begin{bmatrix} P_n & Q_n \\ R_n & S_n \end{bmatrix} \begin{Bmatrix} \bar{c}_{\mathrm{T}n} \\ \bar{c}_{\mathrm{B}n} \end{Bmatrix}$$

where P, Q, and S are the coefficients relating the fluxes and concentrations at the top and bottom of the soil layer. Further details can be found in Rowe and Booker (1985*a*).

Equation [21] represents contaminant transport through the nth horizontal soil layer. By taking into account continuity of concentration and contaminant flux at each layer interface, the description of contaminant transport through a single layer may be assembled into a global matrix representing contaminant transport through the entire soil profile of N layers. This leads to the following set of simultaneous equations:

$$[22] \qquad \begin{Bmatrix} \bar{f}_{\mathrm{T}1} \\ 0 \\ 0 \\ 0 \\ . \\ . \\ . \\ . \\ 0 \\ \bar{f}_{\mathrm{B}N} \end{Bmatrix} = \begin{bmatrix} P_1 & Q_1 & 0 & 0 & 0\,0\,0\,0 & 0 & 0 \\ R_1 & S_1 + P_2 & Q_2 & 0 & 0\,0\,0\,0 & 0 & 0 \\ 0 & R_2 & S_2 + P_3 & Q_3 & 0\,0\,0\,0 & 0 & 0 \\ 0 & 0 & R_3 & S_3 + P_4 & 0\,0\,0\,0 & 0 & 0 \\ . & . & . & . & . \;.\;.\;. & . & . \\ . & . & . & . & . \;.\;.\;. & . & . \\ . & . & . & . & . \;.\;.\;. & . & . \\ . & . & . & . & . \;.\;.\;. & . & . \\ 0 & 0 & 0 & 0 & 0\,0\,0\,0 & P_N + S_{N-1} & Q_N \\ 0 & 0 & 0 & 0 & 0\,0\,0\,0 & R_N & S_N \end{bmatrix} \begin{Bmatrix} \bar{c}_{\mathrm{T}1} \\ \bar{c}_{\mathrm{T}2} \\ \bar{c}_{\mathrm{T}3} \\ \bar{c}_{\mathrm{T}4} \\ . \\ . \\ . \\ . \\ \bar{c}_{\mathrm{T}n} \\ \bar{c}_{\mathrm{B}n} \end{Bmatrix}$$

Equation [22] represents N simultaneous equation in $N + 2$ unknowns. However, introducing appropriate boundary conditions into eq. [22] and solving the system of equations reveals the concentrations at each layer interface. Once this is complete, the contaminant flux at each interface may be calculated using eq. [13]. The boundary conditions for the example problem are described in detail in the next section (see eqs. [25] and [27]).

Illustrative example

For the purposes of demonstrating the proposed analysis method and to illustrate some of the more important effects of contaminant flow through a partially saturated soil liner, a landfill similar to that described by Rowe et al. (1985*a*) is analyzed. It is a waste landfill of considerable lateral extent containing a finite mass of contaminant. The geometric details of the landfill are shown in Fig. 1. The principal components of the landfill from the top down are as follows: (*i*) a volume of waste that is covered by a capping layer; (*ii*) a primary leachate collection system; (*iii*) a geocomposite liner consisting of a geomembrane overlying a natural soil layer (the soil layer is hereafter referred to as the soil

Table 1. Summary of soil parameters used in the illustrative example.

	Soil 1	Soil 2
	Hydraulic properties	
Soil type	Clay	Sand
Soil properties modelled on	Yolo light clay (Moore 1939; Philip 1957)	Typical values
Description	Silt 46%, clay 31%, sand 24%	sand > 95%
Porosity n_c	0.49	0.35
ϑ_{sat}	0.49	0.35
α (m^{-1}; see eq. [1])	0.3	1.0
D_{mo} (m^2/year; see eq.[3])	0.046	0.110
γ (see eq. [3])	11.05	22.26
	Contaminant transport parameters	
Transport parameters based on	Silty clay loam (Rothamsted, in Barraclough and Tinker 1981)	Estimated values
D_{sat}	0.010	0.0186
Λ (see eq. [12])	–0.0044	–0.0064
Ω (see eq. [12])	0.0290	0.0715
ϑ_l	0.15	0.09

liner); (*iv*) a saturated aquifer immediately below the unsaturated soil liner (i.e., the air-entry value of the soil is taken to be zero for simplicity); and (*v*) a layer immediately beneath the aquifer which is impermeable to both water and contaminant.

Moisture profiles through the unsaturated soil liner are evaluated subject to the saturated condition at the interface between the soil liner and the base aquifer ($\vartheta_B = \vartheta_{sat}$, where ϑ_{sat} is the moisture content of the saturated soil), together with a specified Darcy flux through the soil liner. Most geomembranes adopted in practice will have very low permeativity to water. For geocomposite liners constructed under rigorous quality assurance programmes, research by Bonaparte and Gross (1990) has shown that anticipated advective velocities across the geomembrane are very small, ranging from close to 0 mm/year to usually less than 3 mm/year. When a geomembrane liner is installed in the field, small undetected holes may be introduced during seaming. The analysis of contaminant transport through these holes requires an analysis separate from that described here (Rowe 1998).

For the example landfill considered here, the following parameters are adopted:

Landfill	
Landfill length L (m)	200
Height of leachate H_f (m)	1.0
Initial contaminant concentration c_o (kg/m^3)	1.0
Geocomposite liner	
Thickness of geomembrane w_g (mm)	3
Geomembrane mass transfer coefficient D_g (m^2/year)	10^{-2}, 10^{-4}, 10^{-6}, 10^{-8}
Thickness of natural soil liner H (m)	5.0
Free solution diffusion coefficient for contaminant (aqueous solution, infinite dilution, 25°C) D_f (m^2/year)	0.031
Effective porosity of natural soil liner n_c	0.49 (clay liner), 0.35 (sand liner)
Base aquifer	
Thickness of aquifer h_a (m)	1.0
Effective porosity of aquifer n_b	0.3

Contaminant boundary conditions

It is assumed that the maximum concentration above the liner in the landfill (c_{lo}) occurs shortly after closure of the landfill and that this concentration will decrease with time as contaminant moves into the liner–aquifer system. The initial mass of contaminant in the landfill is specified by the parameter H_f, known as the equivalent height of leachate, and is equal to the total volume of leachate at initial concentration c_{lo} divided by the plan area of the landfill.

For the case of a liner without a geomembrane (it is noted that a partially saturated soil may be present without a geomembrane installed, providing the moisture flux through the base of the landfill is sufficiently small), the concentration at the top of the soil liner, $c_{T1}(t)$, is taken to be equal to the concentration in the landfill, $c_l(t)$. The concentration of contaminant in the landfill at some later time is then given by (Rowe et al. 1995)

$$[23] \qquad c_l(t) = c_{T1}(t) = c_{lo} - \frac{1}{H_f}\int_0^t f_{T1}(c,\tau)\mathrm{d}\tau \qquad \text{at } z = 0$$

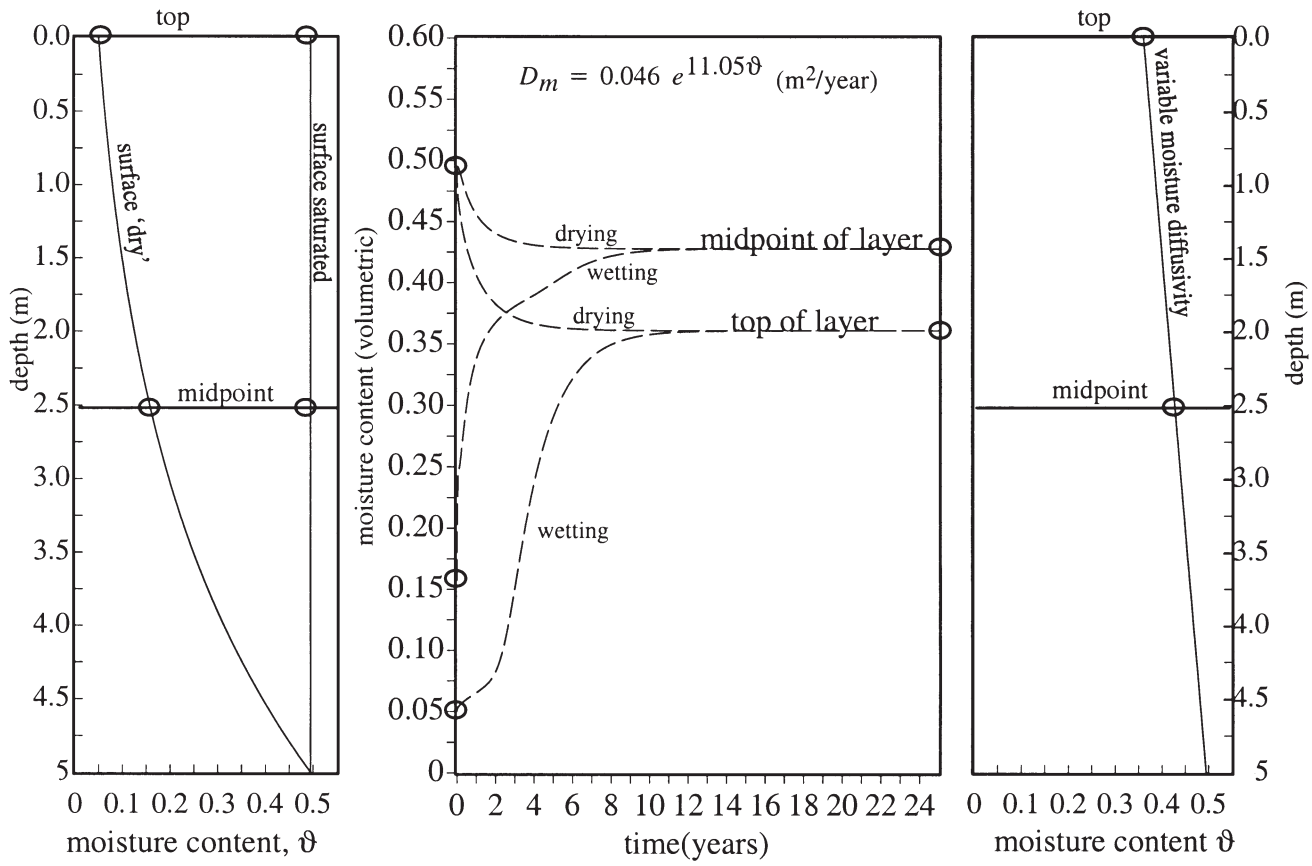
where $f_{T1}(c,t)$ is the contaminant mass flux into the top of the soil liner (i.e., at $z = 0$), and τ is a dummy integration variable.

Rowe et al. (1995) report that a geomembrane in a landfill liner system may be modelled directly as a very thin layer using their semianalytic method. In this paper, an alternate approach is taken. The boundary condition of eq. [23] can be modified to include a geomembrane thickness w_g immediately above the soil liner. In this case, the flux in eq. [23] is defined by the geomembrane properties. Most reported research on the diffusive mass transport through geomembranes is predicated upon the assumption that the mass transfer is proportional to the contaminant concentration difference across the membrane. Hence,

$$[24] \qquad f_{T1}(c,t) = \frac{D_g}{W_g}[c_l(t) - c_{T1}(t)]$$

where D_g is the mass transfer coefficient for the contaminant through the geomembrane. Then, substituting eq. [24] in eq. [23] and taking the Laplace transform leads to

Fig. 2. Moisture-equilibration times in a partially saturated clay profile.



(a) initial moisture content profiles (b) time variation in moisture content at top and midpoint of soil layer (c) final (steady-state) moisture content profile

$$[25] \qquad \bar{f}_{T1} = \frac{c_{lo} - s\bar{c}_{T1}}{s\left(\dfrac{w_g}{D_g} + \dfrac{1}{sH_f}\right)}$$

Equation [25] may now be substituted directly into eq. [22].

We next consider the contaminant behavior at the base of the soil liner. Though truly a two-dimensional problem, it is assumed that the concentration of contaminant in the base aquifer does not vary in the vertical or horizontal direction, and that the contaminant transport in this layer takes place by advection alone. This approximation is considered adequate for many practical situations where the base velocity is relatively small (Rowe and Booker 1985*a*, 1985*b*). The net change in the mass of contaminant in the base aquifer at any time t will be equal to the difference between the contaminant mass flux entering the aquifer from above and the contaminant mass flux leaving the aquifer from the side. It follows that the aquifer concentration $c_{BN}(t)$ is given by

$$[26] \qquad c_{BN}(t) = \int_0^t \frac{f_{BN}(c,\tau)}{n_b h}\,d\tau - \int_0^t \frac{v_b c_{BN}}{n_b L}\,d\tau \qquad \text{at } z = H$$

where $f_{BN}(c,t)$ is the contaminant mass flux into the aquifer from the soil liner above, and v_b is the advection in the base aquifer. Laplace transforming eq. [26] and rearranging leads to

$$[27] \qquad \bar{f}_{BN} = sn_b h_a \bar{c}_{BN}\left(1 + \frac{v_b}{sn_b L}\right)$$

Equation [27] may also be substituted directly into eq. [22]. Following substitution of eqs. [25] and [27] into eq. [22], eq. [22] now involves N simultaneous equations in N unknowns, and this system of equations may be solved using standard numerical methods.

Material parameters for soil liner

Two soil types are considered for the unsaturated soil liner immediately beneath the geomembrane and above the aquifer. Where possible the soil parameters are based on those of real soils, however, it is noted that the soil properties in Table 1 do not represent the estimated behavior of any one soil, but rather represent a collage of typical values for soils of each type. To the authors' knowledge, there is no reported research giving all the parameters for a particular soil required for an analysis of contaminant transport through an unsaturated soil liner beneath a landfill.

Fig. 3. Arrangement for verification of steady-state solutions.

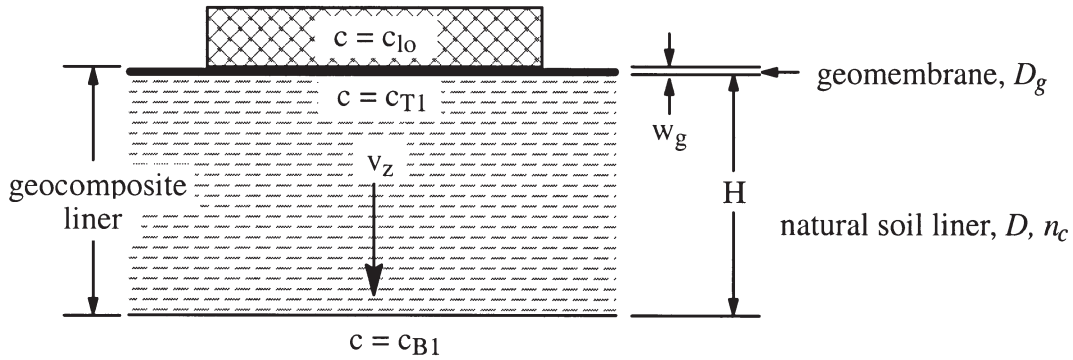
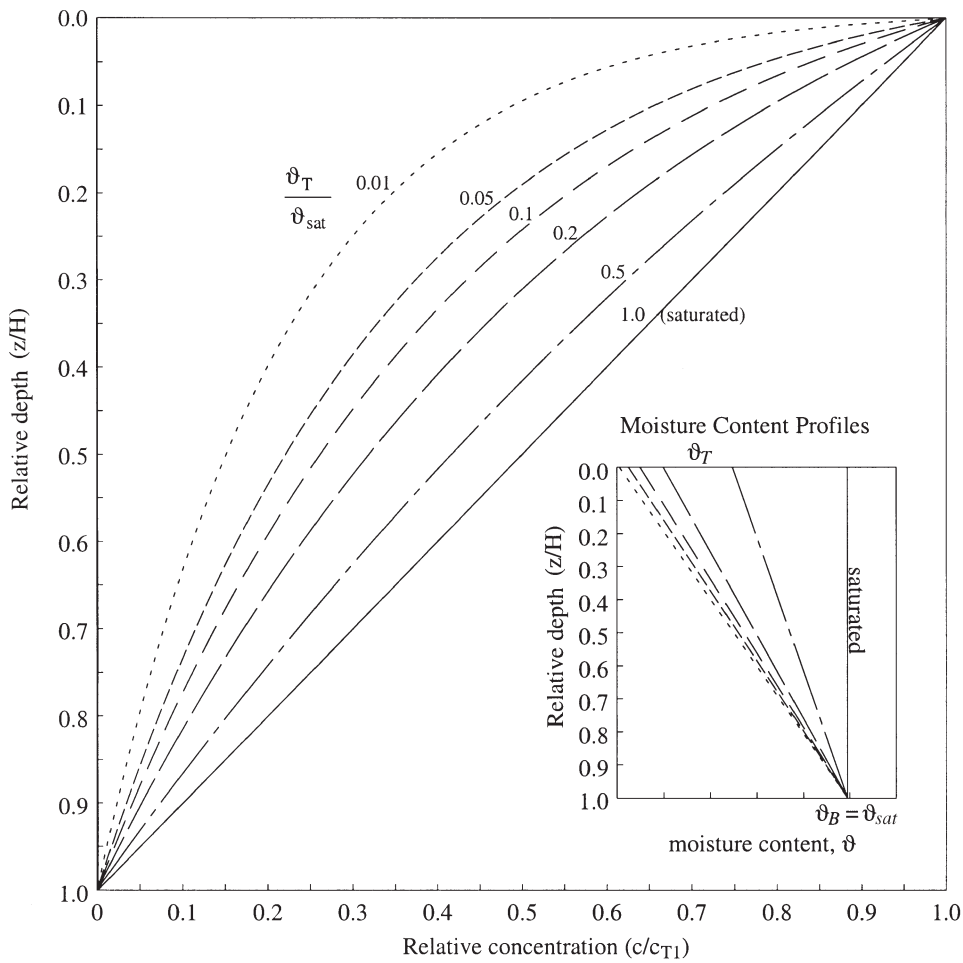


Fig. 4. Comparison of steady-state concentration profiles for differing volumetric moisture content profiles.



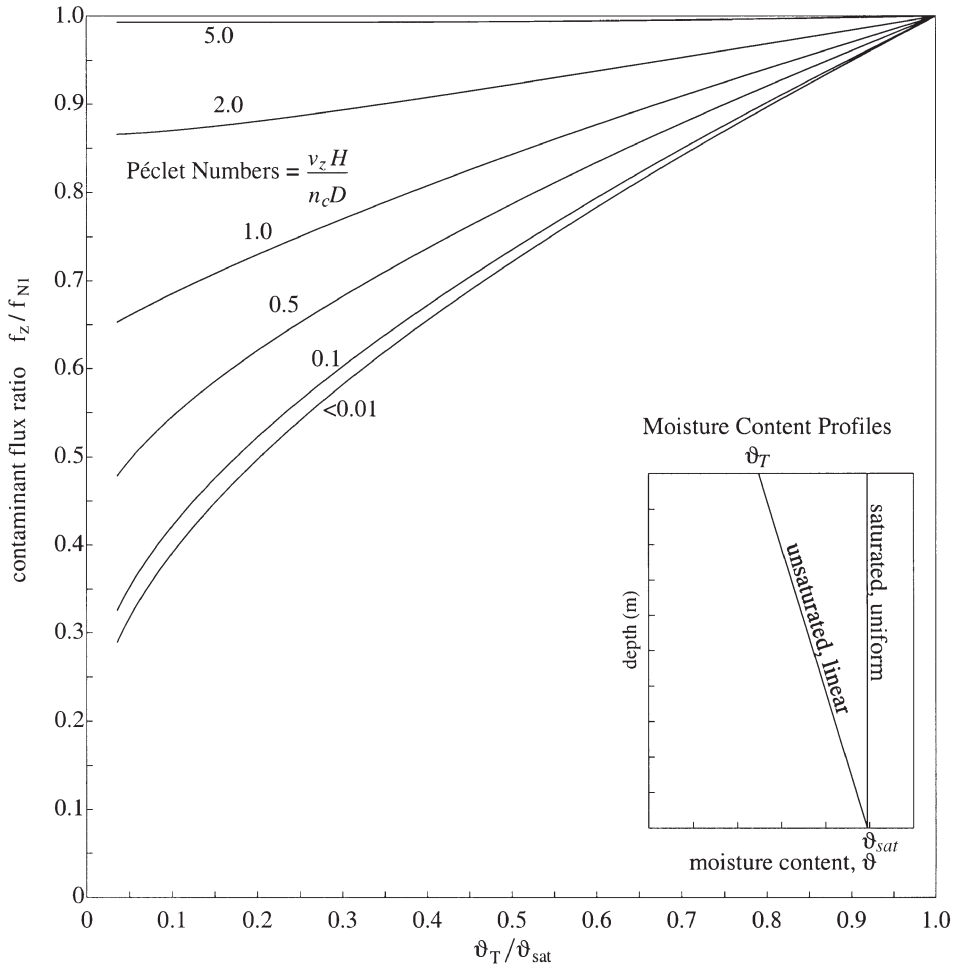
Verification

Numerical verification of the validity of the assumption of steady-state moisture conditions beneath the landfill

The analysis of contaminant transport in this paper is based upon the assumption that the moisture profile beneath the landfill is time invariant. However, it is well known that construction of a covered area over a natural soil will result in transient moisture changes, as infiltration and evapotranspiration from the top of the soil are prevented (Lytton 1969). Therefore, the time required for equilibrium moisture conditions to be established, compared with the time required for contaminant transport through the soil liner, is of importance. It is clear that the coarser the soil, the more quickly equilibrium moisture conditions will be achieved. It can be shown by the solution of Richards equation (eq. [1]) that in the case of the sand soil with parameters shown in Table 1, equilibrium moisture conditions are achieved in a matter of days.

The time required to reach moisture equilibrium for the clay soil, with parameters shown in Table 1, has been investigated by Fityus and Smith (1997). The results are repro-

Fig. 5. Comparison of contaminant mass fluxes through unsaturated moisture profiles, with differing downward advective flow.



duced in Fig. 2, which shows the time rate of change of moisture content at the top of the soil layer and at the mid-height of the soil layer. Both "wetting" and "drying" curves are shown for the soil liner initially "dry" and initially "wet." As expected, both curves asymptote smoothly to the equilibrium moisture condition.

It is clear from the curves, that moisture equilibration in the clay liner is of the order of 8–10 years, and that the majority of the moisture change has occurred within 6–8 years. Given the period of time associated with construction to closure of the landfill (perhaps a decade or more), and the time taken for say 10% of the contaminant mass to traverse the unsaturated soil liner and enter the base aquifer (found to be at least hundreds of years), it is concluded that the steady-state moisture profile beneath the landfill is a reasonable approximation for the analysis of the landfill considered here.

Verification of contaminant transport analysis through an unsaturated soil

As mentioned previously, the differential equation governing contaminant transport through saturated soils is a particular case of the more general contaminant transport through unsaturated soil. By choosing the contaminant transport parameters in eq. [14] to represent a saturated soil, the example problem considered by Rowe and Booker (1985*a*, 1985*b*) was reanalyzed, and it was found that the two analyses were in agreement, at least to within plotting accuracy of the contaminant profiles and mass transfer curves.

As a means of checking the Chebyshev polynomial series solutions to eq. [14] for an unsaturated soil liner, steady-state analytic solutions for contaminant transport through a single, partially saturated, isotropic, homogeneous soil layer were found (see Fig. 3). It was assumed that the diffusion coefficient was constant and that the moisture content varied linearly across the soil layer of thickness H, from saturated at the bottom ($\vartheta_{sat} = \vartheta_B = n_c$), to partially saturated at the top (ϑ_T) (see inset to Fig. 4). For this case the governing differential equation for contaminant transport through the partially saturated soil is

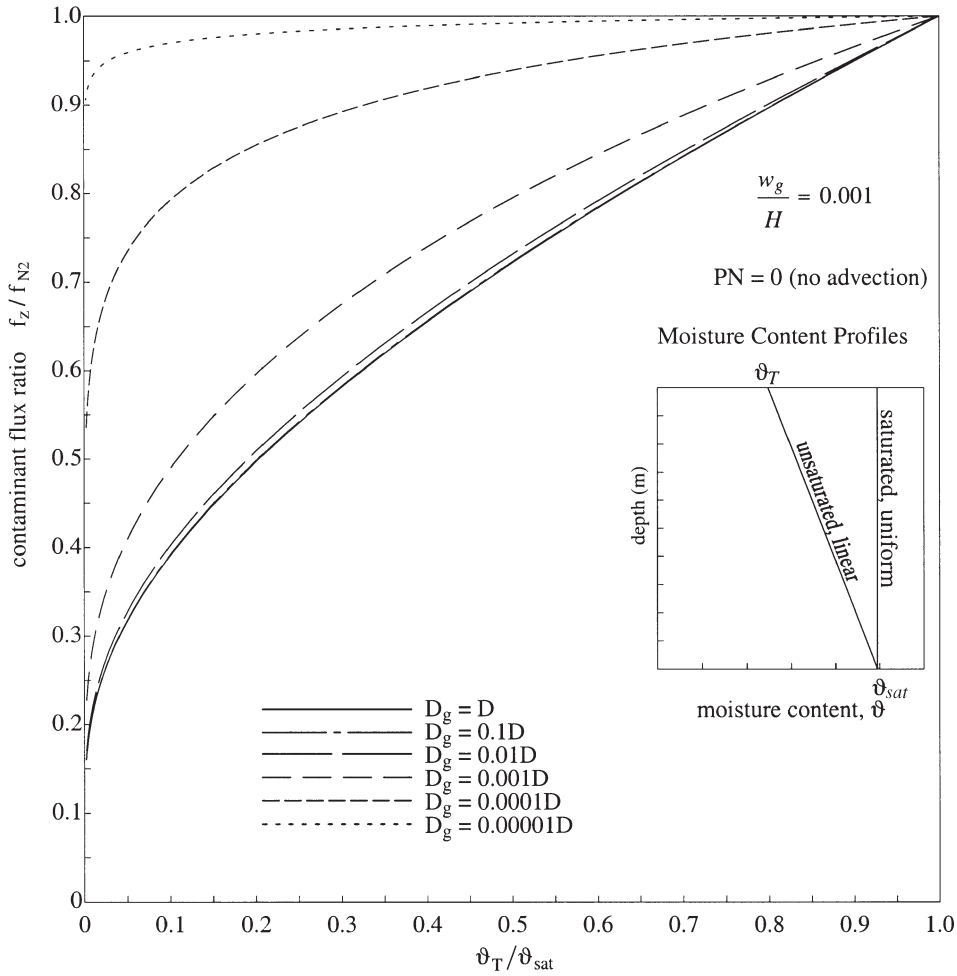
$$[28] \qquad D(A+Bz)\frac{d^2c}{dz^2} + (DB - v_z)\frac{dc}{dz} = 0$$

which has a general solution

$$[29] \qquad c = C_1 + C_2(A+Bz)^{v_z/DB}$$

where C_1 and C_2 are constants that may be evaluated from the boundary conditions. For simplicity, the concentration at the top of the soil liner is taken to be a constant (c_{T1}) and the concentration at the bottom of the soil liner is taken to be zero. It is noted in passing that for the case of zero advection, the solution to eq. [28] reduces to

Fig. 6. Comparison of contaminant mass fluxes through a partially saturated geocomposite liner for various geomembrane diffusion coefficients (D_g).



[30] $$c = C_1 + C_2 \ln(A + Bz)$$

A convenient Péclet number for the soil liner may be defined as PN = $v_z H/n_c D$. Figure 4 shows nondimensional steady-state contaminant profiles through the soil liner for Péclet number zero (i.e., zero advection), and Fig. 5 shows nondimensional steady-state contaminant mass fluxes through the soil liner for a range of Péclet numbers. In Fig. 5 the contaminant fluxes through the soil liner have been normalized with respect to the contaminant mass flux through the soil liner when it is saturated, viz.,

[31] $$f_{N1} = \frac{c_{T1} v_z e^{(v_z H/n_c D)}}{e^{(v_z H/n_c D)} - 1}$$

The steady-state analytic solutions for each Péclet number and the long-term solutions found using the Chebyshev polynomial series solution were found to be in exact agreement, to at least three significant figures.

Analytic solutions for the steady-state contaminant flux through a geocomposite liner with no advection (i.e., a soil liner covered by a geomembrane) were also found. If the contaminant flux through the geomembrane (eq. [24]) is equated with the flux through the soil liner, then the steady-state concentration profile through the geocomposite can be shown to be

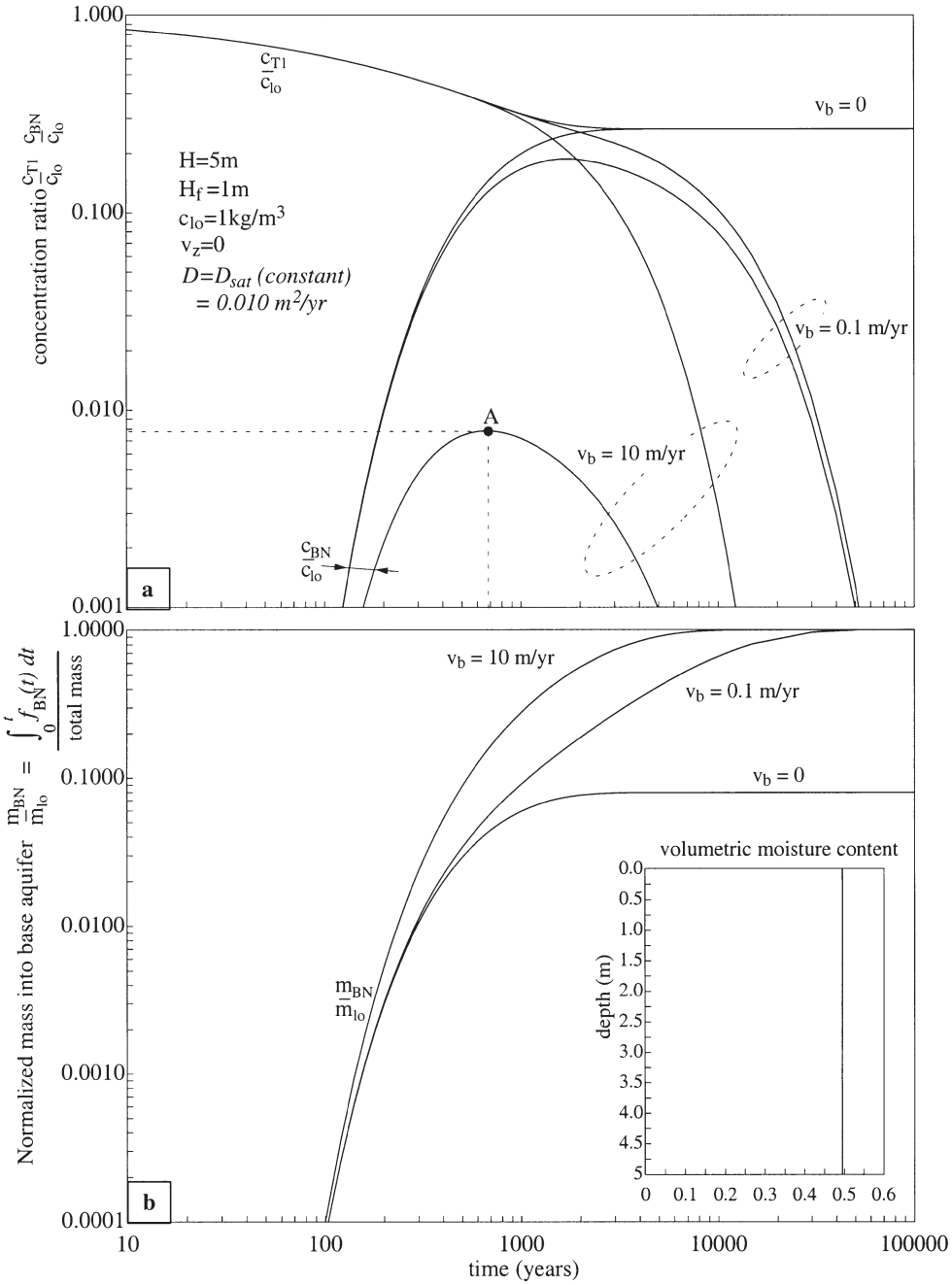
[32] $$c(z) = \frac{c_{lo} D_g \ln(A + BH)}{BDw_g + D_g[\ln(A + BH) - \ln(A)]} + \frac{c_{lo} D_g}{BDw_g + D_g[\ln(A + BH) - \ln(A)]} \ln(A + Bz)$$

and the steady-state flux through the geocomposite liner is

[33] $$f(z) = \frac{c_{lo} BDD_g}{BDw_g + D_g[\ln(A + BH) - \ln(A)]}$$

Figure 6 shows nondimensional steady-state contaminant fluxes through the soil liner for a range of different ratios of the membrane transfer coefficient (D_g) to soil liner diffusion coefficient (D). In Fig. 6 the ratio of the geomembrane thickness (w_g) to soil liner thickness (H) is chosen to be 0.001, and the contaminant flux has been normalized with respect to the contaminant flux through the geocomposite when the soil liner is completely saturated, viz.,

Fig. 7. Contaminant transport solutions for a saturated soil liner with a constant contaminant diffusion coefficient: clay soil.



$$[34] \qquad f_{N2} = \frac{n_c c_{lo} D D_g}{n_c D W_g + D_g H}$$

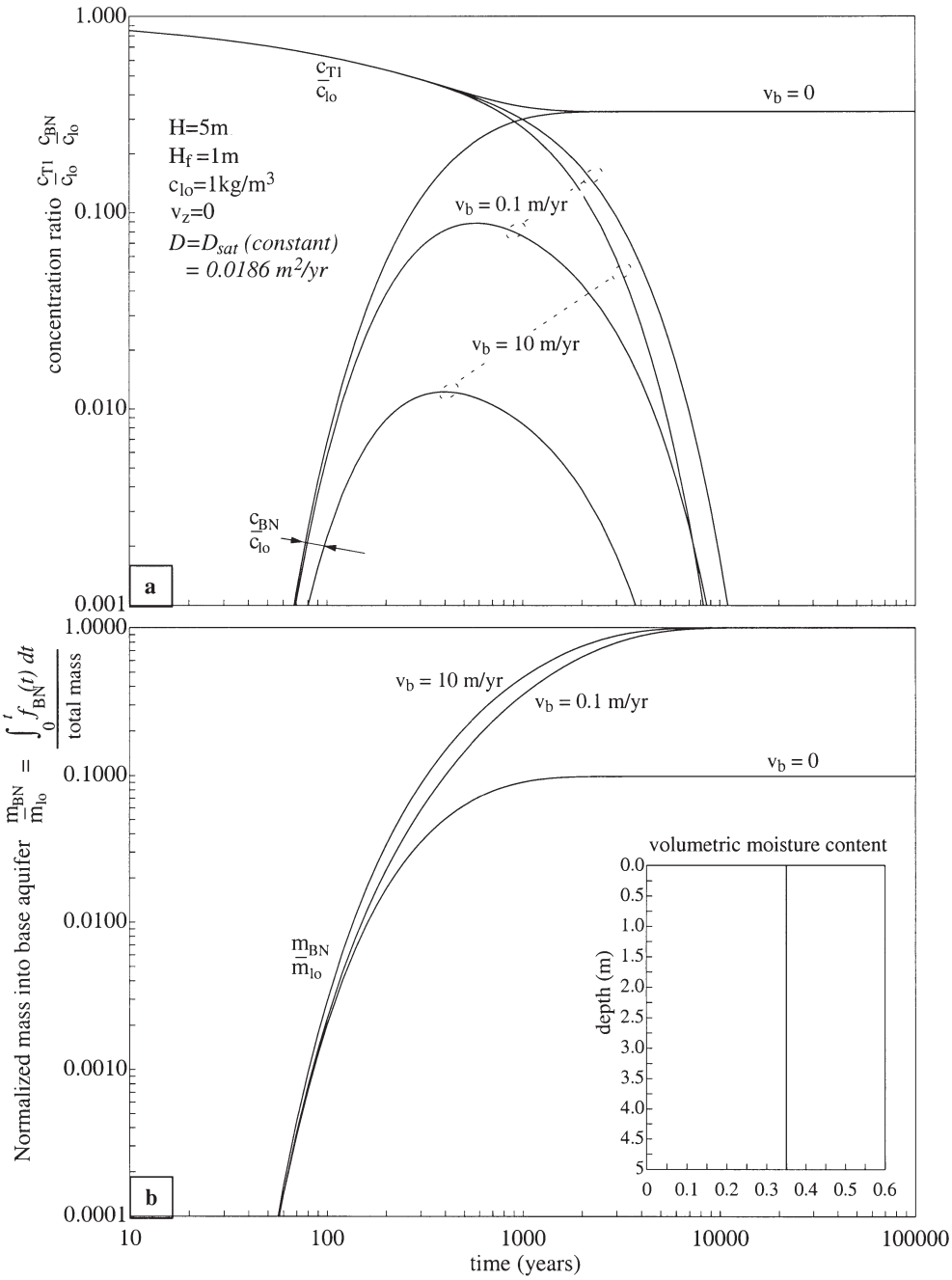
Once again, the steady-state analytic solutions and long-term solutions found using the Chebyshev polynomial series expansion were found to be in exact agreement.

Contaminant transport analysis of the landfill liner

Introduction

In order to have a meaningful reference point for assessing contaminant transport through a partially saturated geocomposite liner, transport through a saturated soil liner, with no geomembrane present, was investigated first. This analysis is referred to as case 1. The next two analyses, also with no geomembrane present, but now with the soil liner partially saturated, are referred to as cases 2 and 3. Cases 2 and 3 are compared and contrasted to case 1. In this way, some of the practical implications of unsaturated soil conditions on contaminant transport through the soil liner may be highlighted. Next, the influence of advection through a soil liner is examined. Finally the mass transfer characteristics of a geocomposite liner are considered, and this behavior is compared and contrasted to the mass transfer characteristics of a partially saturated soil liner. It is noted here that for the sake of brevity, the influence of advection and the

Fig. 8. Contaminant transport solutions for a saturated soil with a constant contaminant diffusion coefficient: sand soil.



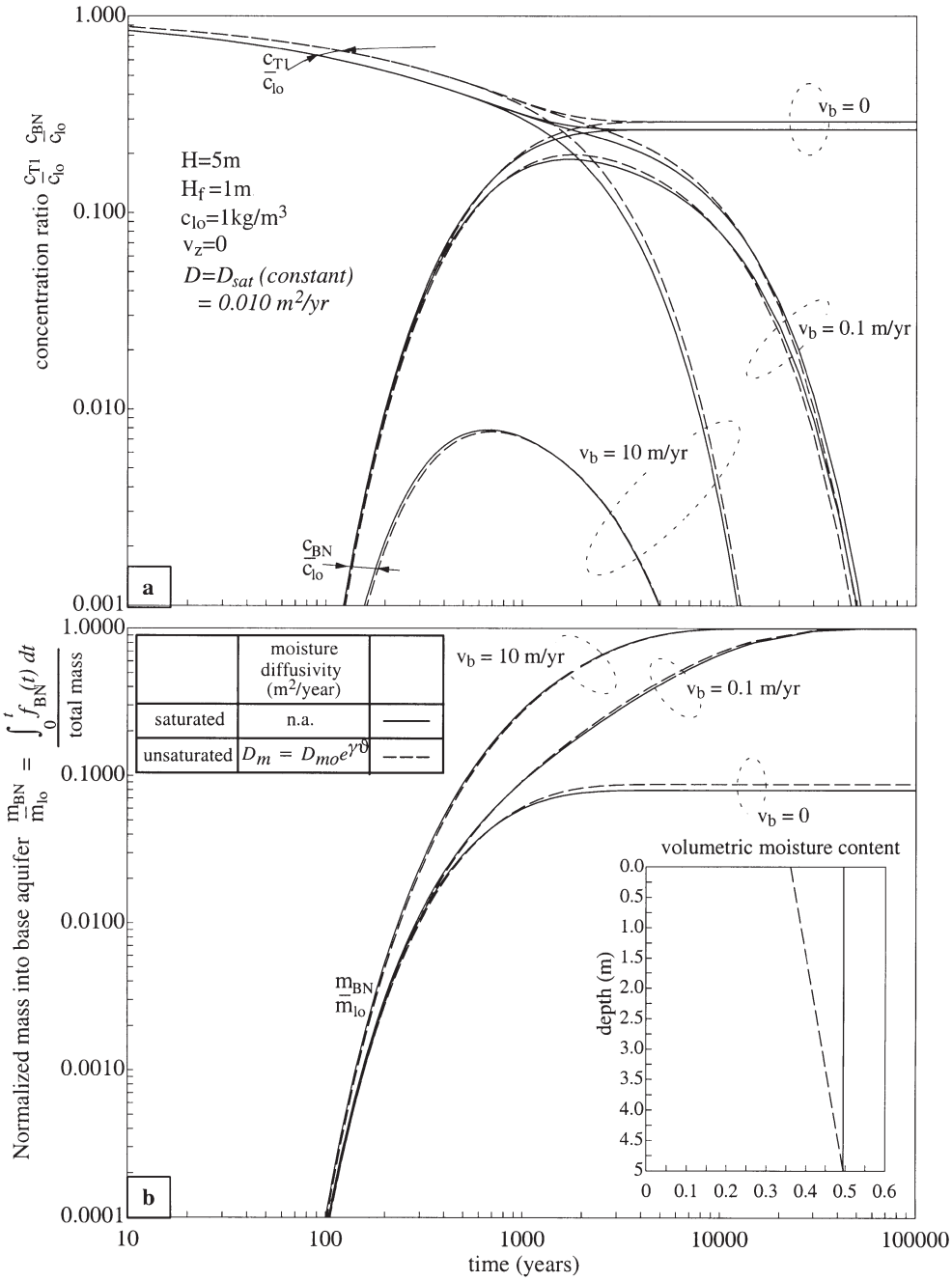
geomembrane on liner performance is only considered for the clay soil liner. We proceed to described cases 1–3 in more detail.

Cases 1–3 examine the response of the soil liner alone (i.e., with no geomembrane present), and with diffusive mass transport being the sole transport mechanism (i.e., with advective transport zero). Three different states of the soil liner are considered as follows: (1) diffusive mass transport through a saturated soil liner with a constant contaminant diffusion coefficient; (2) diffusive mass transport through an unsaturated soil liner, with a constant contaminant diffusion coefficient value, being the same as that for case 1, and with the purpose of case 2 being to allow consideration of the influence of a spatially varying moisture content, without including the effects of moisture dependence of the diffusion coefficient; and (3) diffusive mass transport through an unsaturated soil liner, with the contaminant diffusion coefficient set to vary with the volumetric moisture content.

Case 1: contaminant transport through a saturated soil liner

If the advection in the base aquifer (v_b) is zero, then no mass can escape from the liner–aquifer system and eventually a uniform concentration will be established throughout the soil liner and base aquifer. This is seen to be true for the clay soil liner in Fig. 7 and the sand soil liner in Fig. 8. In each figure, the curve representing the concentration at the top of the soil liner decreases smoothly over time and joins

Fig. 9. Comparison of contaminant transport solutions for saturated and unsaturated soils with a constant contaminant diffusion coefficient: clay soil.



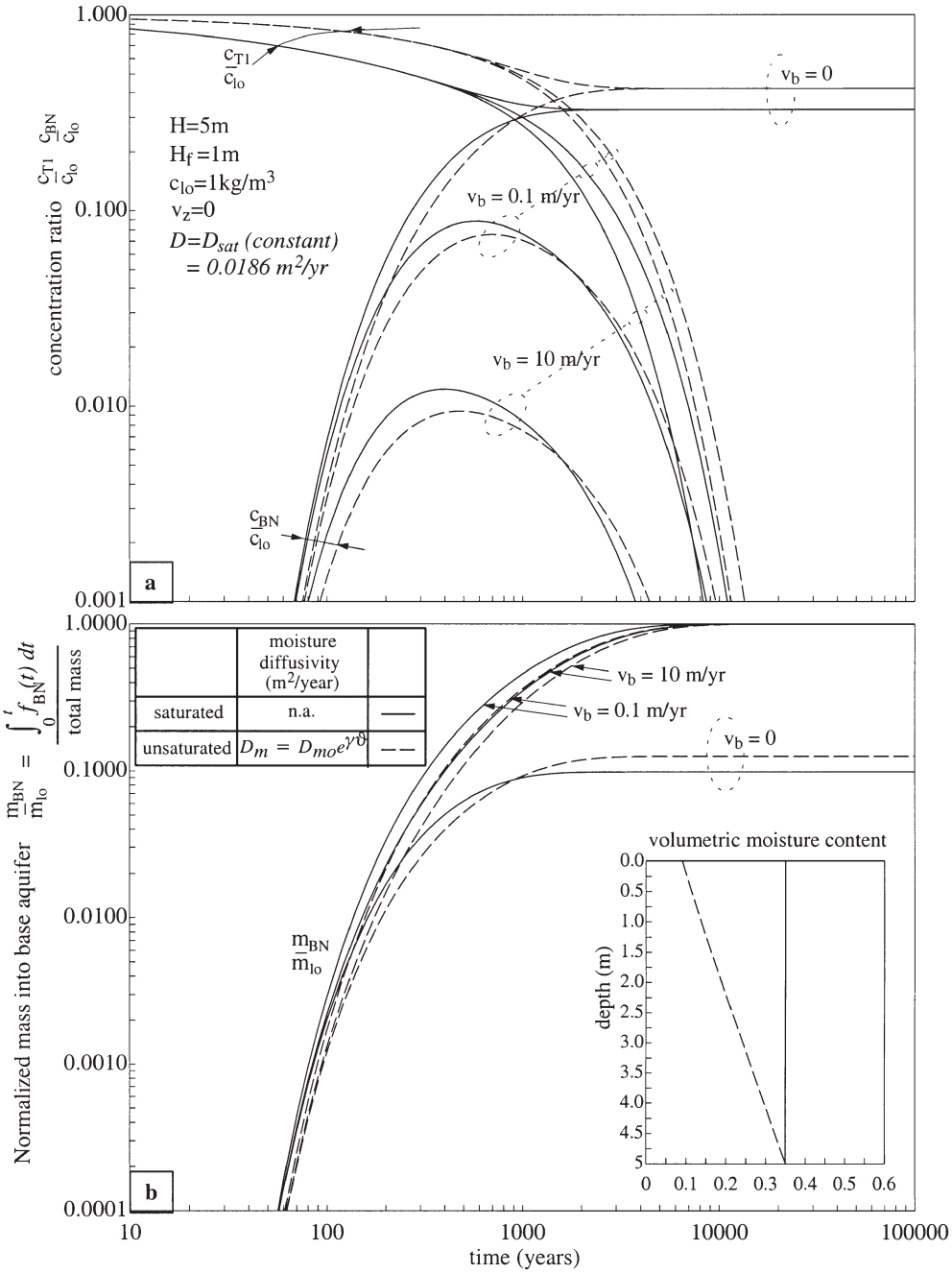
the smoothly increasing curve representing the concentration in the base aquifer. For the clay soil, Fig. 7*a* shows that concentration equilibrium is established in approximately 3500 years, whereas for the sand soil Fig. 8*a* shows that equilibrium conditions are established in approximately 2000 years. The difference in equilibrium times may be attributed to the difference in the diffusion coefficients for the two soil types.

When there is a horizontal advection (v_b) through the base aquifer, contaminant mass is lost from the liner–aquifer system and the long-term concentrations in the landfill and the base aquifer must be zero. This expected behavior is confirmed by examination of Figs. 7*a* and 8*a*. It is also confirmed by Figs. 7*b* and 8*b* where the total contaminant mass flux through the base aquifer, normalized with respect to the initial mass, is seen to be equal to one after a long period of time.

As the horizontal advection through the base aquifer increases, the peak contaminant concentration in the base aquifer is reduced. For the clay soil, when v_b = 0.0 m/year, the peak nondimensional concentration in the base aquifer is approximately 0.2, and this decreases to 0.008 when v_b increases to 10.0 m/year.

We here define "breakthrough" of the contaminant through the soil liner to mean the time required for 0.01% of the initial mass in the landfill to pass through the soil liner into the base aquifer. It is seen from Fig. 7*b* that this occurs

Fig. 10. Comparison of contaminant transport solutions for saturated and unsaturated soil with a constant contaminant diffusion coefficient: soil 2, sand soil.



for the clay soil after 100 years, whereas for the sand soil Fig. 8*b* shows that breakthrough occurs after 60 years.

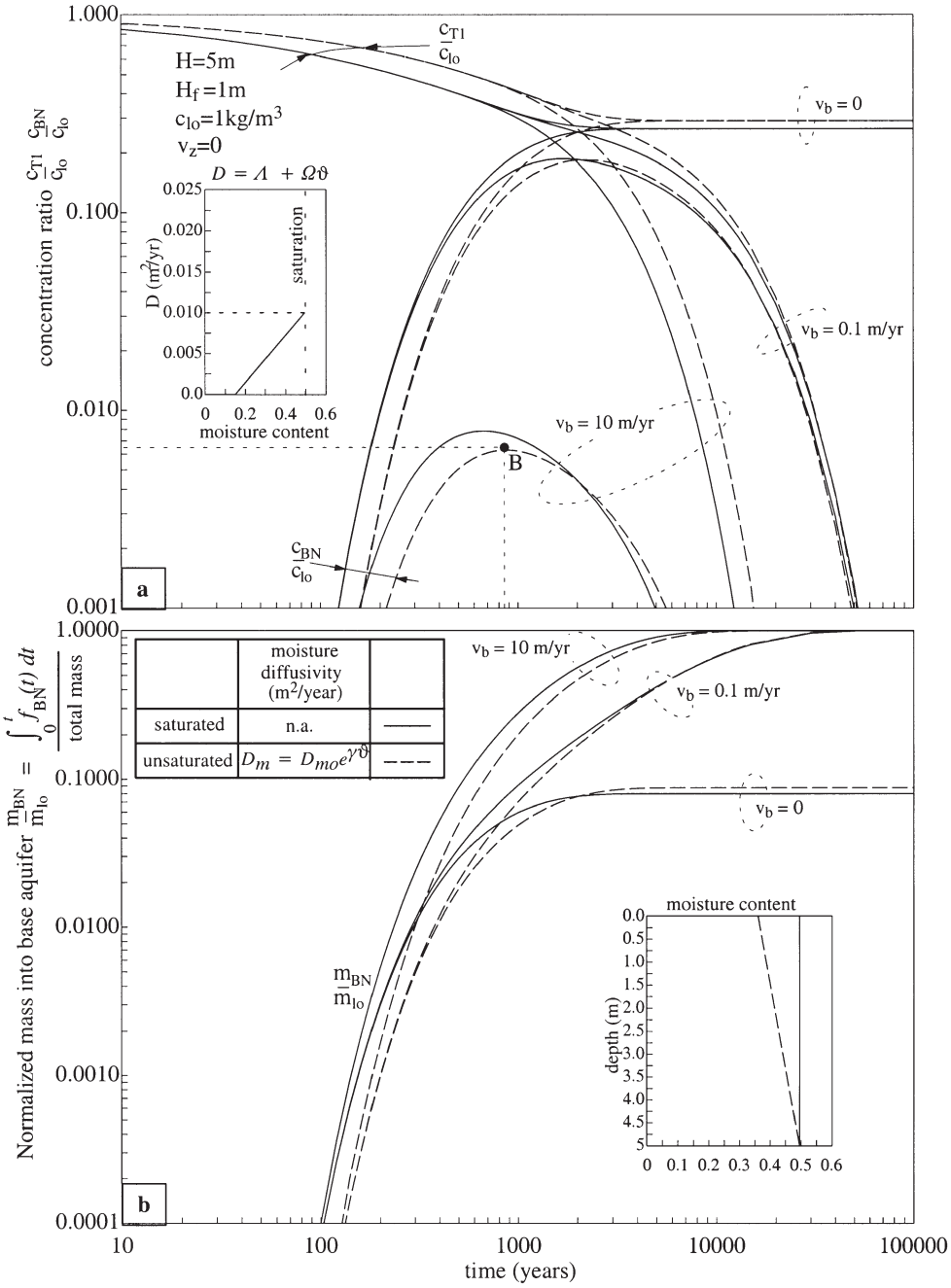
In this case, both the sand and clay soils were saturated under conditions of zero advection. Practically this could be achieved in a number of different ways, but the simplest is to raise the water table to the top of the soil liner.

Case 2: contaminant transport through an unsaturated soil liner with a constant diffusion coefficient

For this analysis, the conditions are the same as those for case 1 except that the soil liner is now unsaturated, with the moisture profile calculated according to eq. [6]. Moisture profiles within the unsaturated soil liner are evaluated subject to a saturated boundary condition at the contact with the base aquifer ($\vartheta_B = \vartheta_{sat}$, i.e., water table at the base of the soil liner) and zero Darcy flux through the soil liner. The air-entry value of the soil is taken to be zero for simplicity, although nonzero values may be easily accommodated in the analysis.

For the clay soil, the decrease in moisture content at the top of the soil liner is relatively small. The inset to Fig. 9*b* shows the volumetric moisture content at the top of the soil liner is 0.36, with ϑ_{sat} = 0.49 at the bottom of the soil liner. The ratio of the decrease in moisture content at the top of the clay soil liner to the saturated moisture content is about 26%. For the sand soil, the inset to Fig. 10*b* shows a mois-

Fig. 11. Comparison of contaminant transport solutions for a saturated and unsaturated soil with a variable contaminant diffusion coefficient: clay soil.



ture content at the top of the soil liner of 0.10, with ϑ_{sat} = 0.35 at the bottom of the soil liner. The ratio of the decrease in moisture content at the top of the sand soil liner to the saturated moisture content is about 71%.

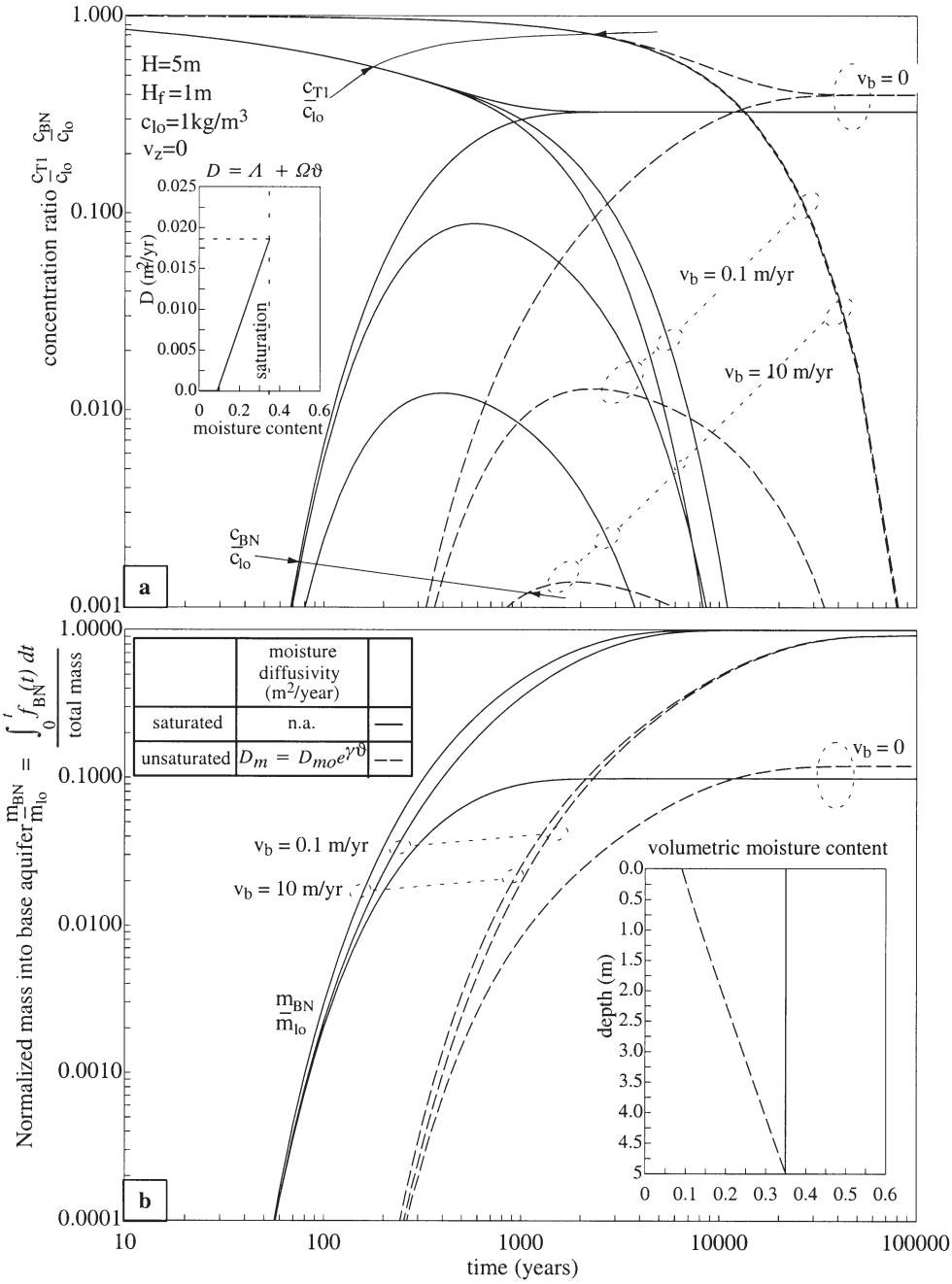
Figures 9 and 10 show the contaminant transport solutions for case 1 as solid lines, and those for case 2 as broken lines.

For the clay soil liner, it is apparent that the cases 1 and 2 contaminant transport solutions are nearly the same, despite the 26% reduction in moisture content at the top of the liner in case 2. For the sand soil the contaminant transport solutions for cases 1 and 2 differ more, but not greatly so. This is despite the 71% reduction in moisture content at the top of the sand liner in case 2. Clearly, the reduction in the cross-sectional area available for contaminant mass transport at the top of the soil liner is not having the significant influence on the contaminant mass transfer through the soil liner that may be expected intuitively. The reasons for this behavior are examined in detail in the discussion.

Case 3: contaminant transport through an unsaturated soil liner with a variable diffusion coefficient

For case 3, the conditions are the same as those for case 2 except that the diffusion coefficient is dependent on the volumetric moisture content. Figures 11 and 12 again show the contaminant transport solutions for case 1 as solid lines and those for case 3 as broken lines.

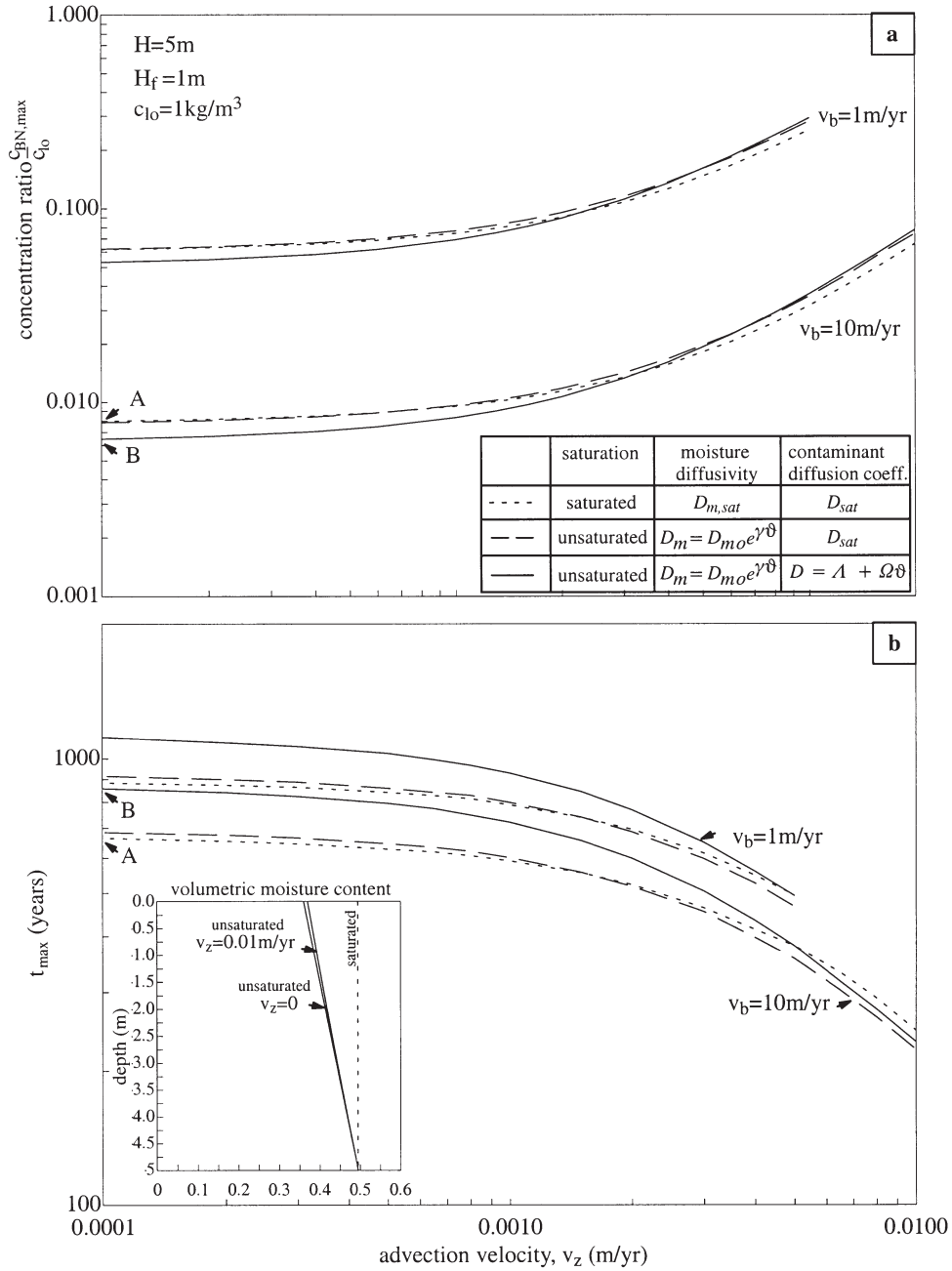
Fig. 12. Comparison of contaminant transport solutions for a saturated and unsaturated soil with a variable contaminant diffusion coefficient: soil 2, sand soil.



Clearly if there is no contaminant mass loss from the liner–aquifer system, the final contaminant concentrations depend only on the total volume of pore fluid in the system. This is true because the equations governing the moisture distribution are independent of equations governing the contaminant behavior. A comparison of Figs. 9 and 10 (case 2) with Figs. 11 and 12 (case 3) shows that this is reflected in the calculated contaminant transport solutions. However, examination of Figs. 11 and 12 shows that moisture dependence of the diffusion coefficient does have a pronounced effect on the rate at which contaminant moves throughout the soil liner. For example, the time required for the liner–aquifer system to reach concentration equilibrium is significantly increased for case 3 compared with case 2. For the clay soil, equilibrium times are increased from 3500 to 5000 years, and for the sand soil from 2000 to 40 000 years. There is also a significant delay in the breakthrough time for both liners, increasing from 100 to 200 years for the clay liner and from 60 to 250 years for the sand liner.

When there is contaminant mass loss from the liner–aquifer system due to the presence of a horizontal velocity in the pore fluid of the base aquifer, the predicted time for the entire contaminant mass to pass through the system is increased considerably if the diffusion coefficient is moisture dependent. A comparison of case 1 solutions and case 3 solutions shows that the time for the entire contaminant mass to

Fig. 13. Contaminant transport analysis with vertical advective flow: clay soil.



pass through the liner–aquifer system increases from a few thousand years to greater than 10 000 years for both the clay and sand liners.

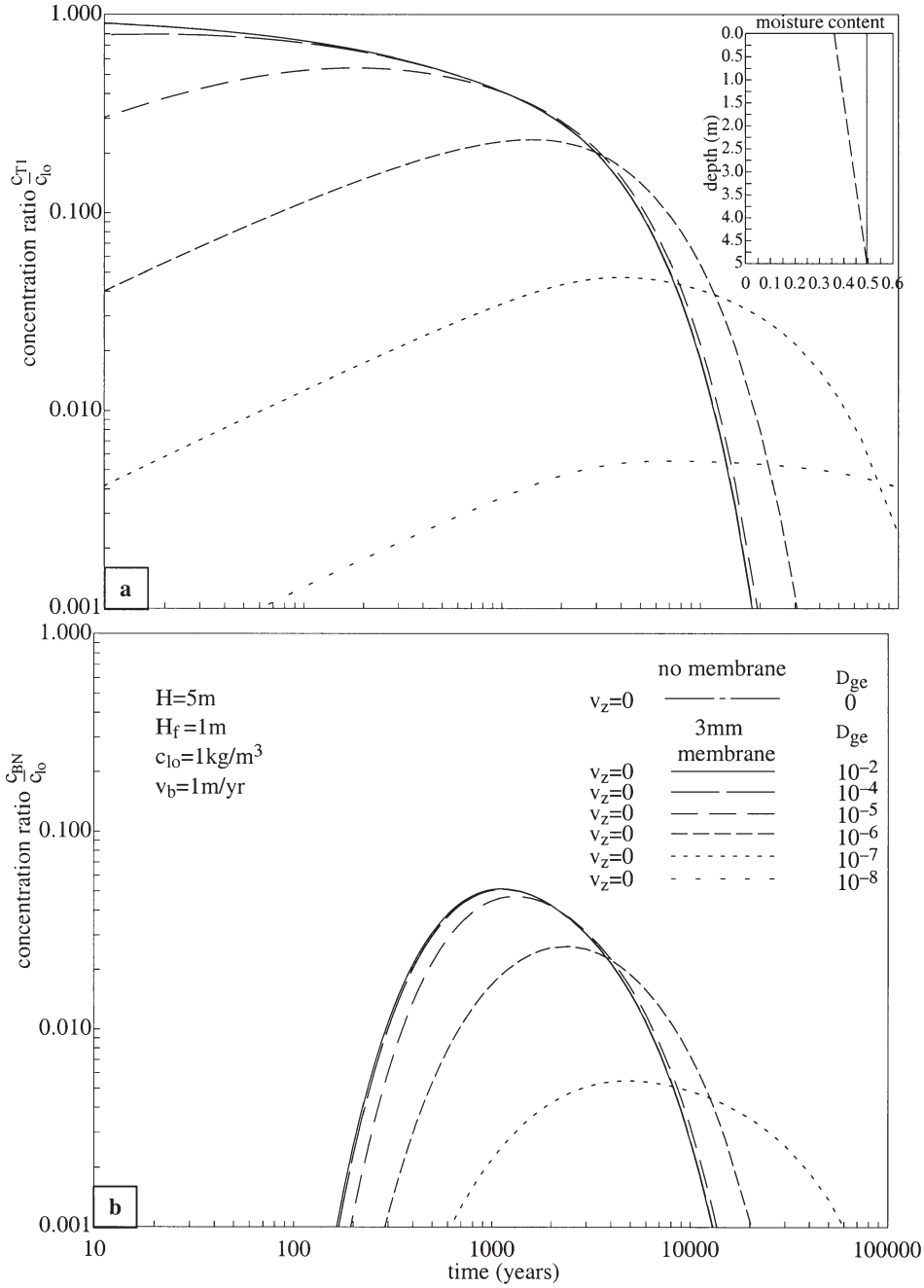
Influence of advection on mass transfer through the soil liner

There may be a flux of water through the landfill liner, with the magnitude of the flux depending on many factors. An estimate of the water flux through the liner may be made on the basis of a water mass balance for the whole landfill, or by measurement. Providing the Darcy velocity is not too large, the soil liner will remain partially saturated.

Figure 13 illustrates the influence of advective flow on contaminant transport through the clay soil liner. Apart from the advection, the liner considered here is the same as that considered in case 3. The contaminant transport solutions for the unsaturated soil liner, represented by broken lines, are again compared with the case 1 solutions represented by solid lines. Note, from the inset to Fig. 13*b*, that a vertical Darcy velocity through the liner of 0.01 m/year has only a minor influence on the estimated steady-state moisture profile when compared with the moisture profile for zero advection.

Figure 13*a* shows the effect that a vertical advection has on the maximum contaminant concentration at the base of the liner. It is seen that for advections less than 0.001 m/year, advection has no influence on the peak concentration in the base aquifer. As advection increases above 0.001 m/year, the peak concentration in the base aquifer increases. Of particular relevance is the result that a small

Fig. 14. Contaminant transport solutions for different geomembrane properties: clay soil.



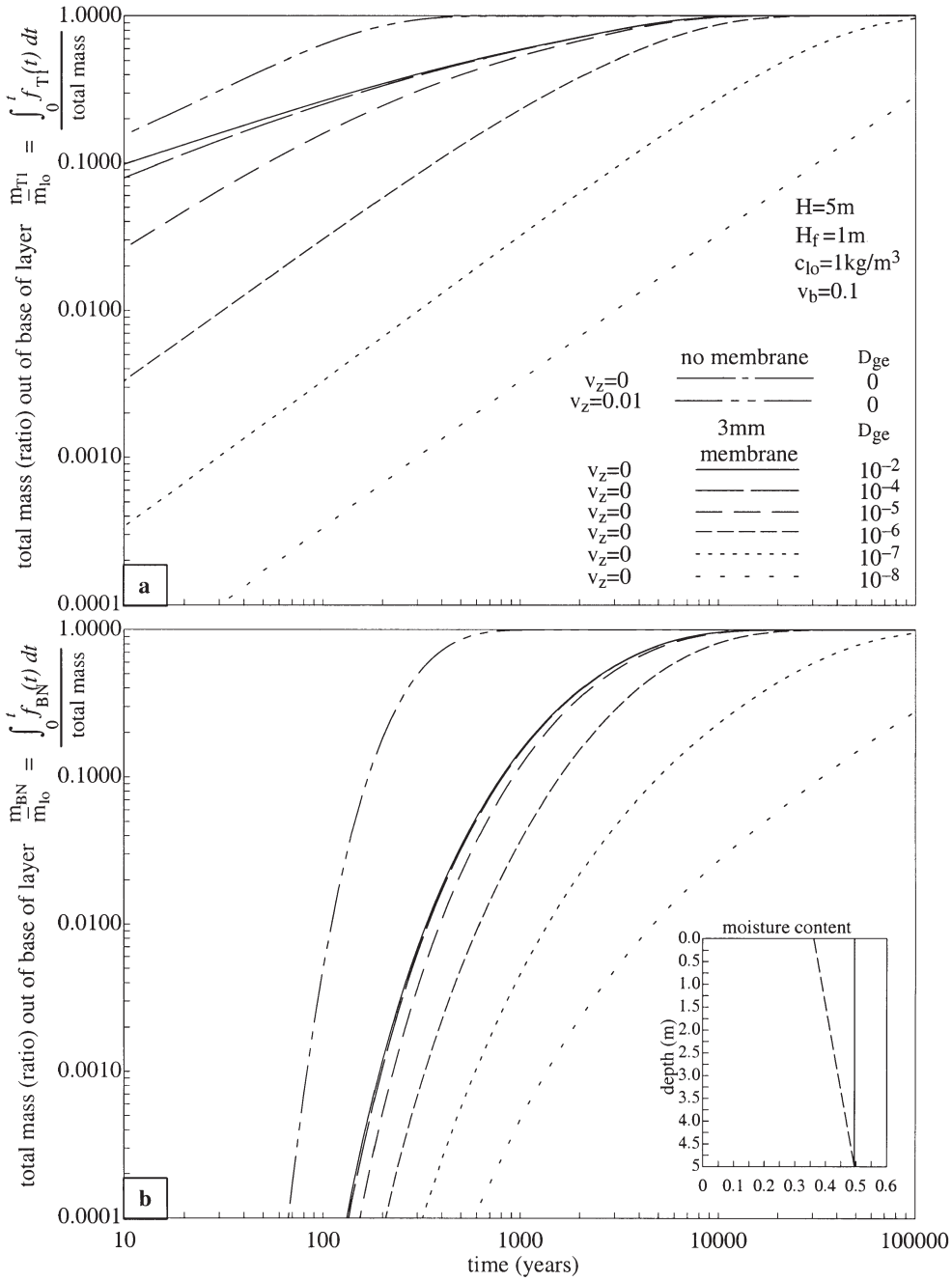
advective flow can substantially increases the contaminant mass flux through the soil liner. For example, an advective flow of just 0.01 m/year results in an order of magnitude increase in the peak concentration observed in the aquifer. This finding highlights the need for effective control of advection. Figure 13*b* shows that as the advection increases above 0.001 m/year, the time at which the peak concentration occurs in the base aquifer decreases. Note that points A and B in Figs. 7 and 11, respectively, are identified in Fig. 13*a*.

Influence of a geomembrane on mass transfer through the landfill liner

Many modern landfill liners are geocomposite liners, consisting of a geomembrane in direct contact with a soil liner. Excluding transport through holes in the geomembrane (which requires a separate analysis), the movement of water through the geomembrane is controlled by the gradient of the Gibb free energy across the geomembrane (Fityus and Smith 1998). Water diffuses across the intact geomembrane, and then causes bulk fluid flow in the soil pore water. Based on a consideration of factors discussed in Giroud and Bonaparte (1989) and Bonaparte and Gross (1990) and recent estimates of the mass transfer characteristics of HDPE liner material (Rowe 1998), it is estimated that the average advective flow through the soil layer of a geocomposite liner would be less than 0.001 m/year.

For the liner analyzed in this section, a 3 mm thick geomembrane is placed on top of the soil liner, but other-

Fig. 15. Contaminant transport solutions for different geomembrane properties: clay soil.

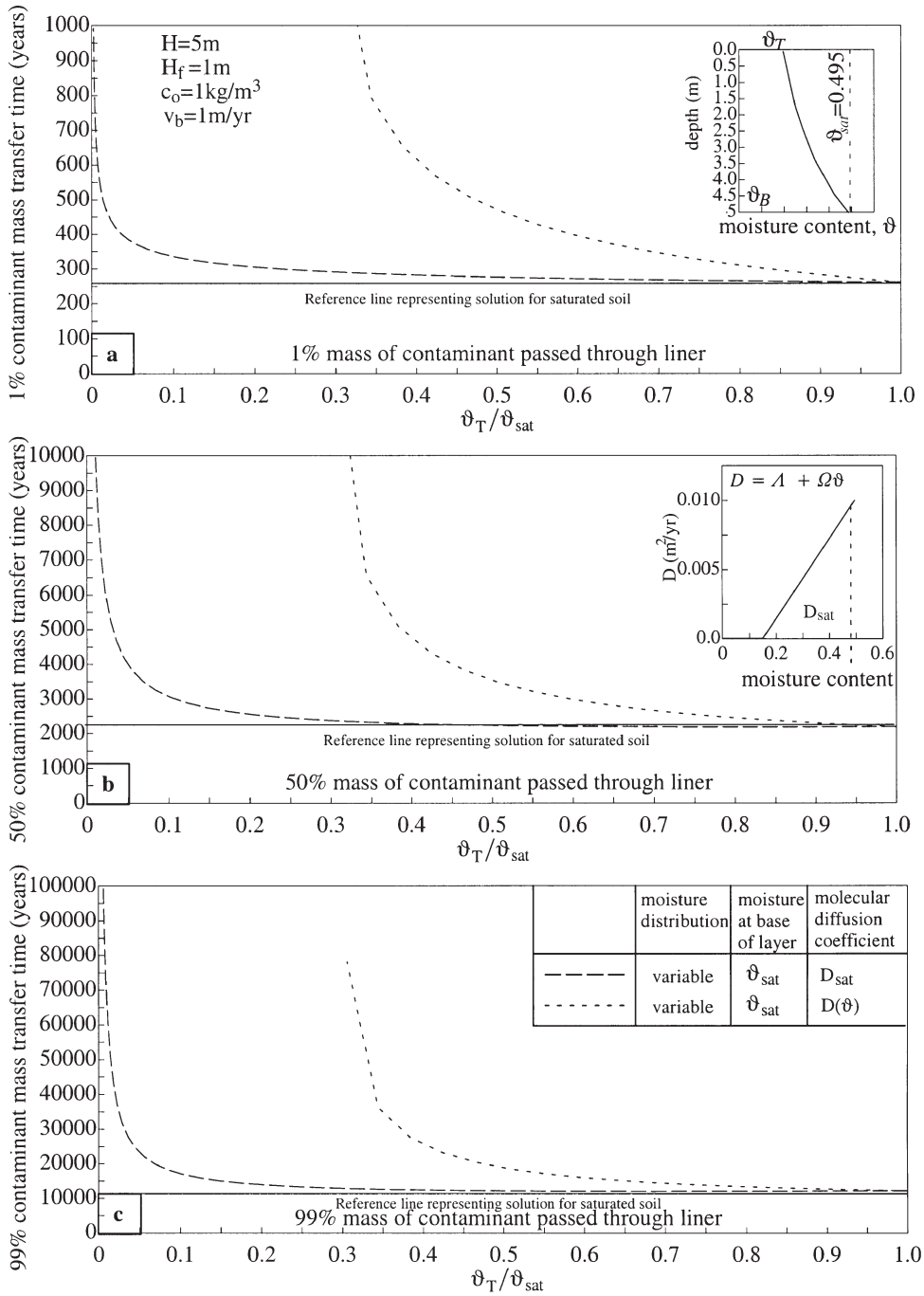


wise the problem variables have the same values as those for case 3; v_b = 1 m/year. As the analysis preformed in the previous section indicated that contaminant mass transfer through the soil liner was unaffected by advective flows less than 0.001 m/year, advective flow in the vertical direction is here taken to be zero.

The time dependence of the normalized concentration at the top and bottom of the soil liner is shown in Fig. 14 for various membrane diffusivities. It is noted that c_{T1} is the concentration immediately beneath the geomembrane (i.e., at the top of the soil liner) and that it is normalized with respect to c_{lo}, the initial concentration in the landfill (i.e., immediately above the geomembrane). Figures 14 and 15 clearly show that for membrane diffusivities greater than 10^{-4} m^2/year, the geomembrane has little effect on the mass transfer characteristics of the liner. For membrane diffusivities less than 10^{-4} m^2/year, the geomembrane has the effect of reducing concentrations throughout the soil liner, delaying contaminant breakthrough time, reducing peak contaminant concentrations in the base aquifer, and delaying the time to reach the peak concentrations in the base aquifer.

A review of research results presented by Rowe (1998) suggests the diffusivity of a HDPE liner is around 10^{-5} m^2/year for a range of contaminants. Examination of Fig. 14*a* shows that a 3 mm thick geomembrane, with a diffusivity for a contaminant of 10^{-5} m^2/year, causes as substantial reduction in contaminant concentration immediately beneath the geomembrane at very early times in the contaminating lifetime

Fig. 16. Comparison of times for the transport of a percentage of contaminant mass through a soil liner: clay soil.



of the landfill (i.e., at the top of the soil liner the nondimensional concentration is 0.3), but only a very small reduction at later times. Figure 14*b* also shows that a HDPE geomembrane 3 mm thick will only have a small influence on the peak concentration in the base aquifer, these observations suggesting that the primary role of the geomembrane in this case is controlling the hydraulic conditions of the liner, rather than preventing contaminant transport. These observations could influence selection of the geomembrane thickness and type.

For the example problem with an unsaturated clay liner and a moisture-dependent diffusion coefficient, it is concluded that a 3 mm thick HDPE geomembrane will confer little benefit on the expected contaminant mass transfer characteristics of the liner, providing advection is controlled. The role of the geomembrane in controlling contaminant transport through the liner would be reduced even further if the soil liner further desaturates, or if the geomembrane were thinner. This contrasts with the more significant benefits conferred for controlling contaminant transport when the

clay liner is saturated.

Discussion of the contaminant transport analysis for the landfill liner

A comparison of the contaminant transport solutions for cases 1 and 2 has shown only minor differences, and it was noted that this may not have been expected intuitively. To remind the reader, both case 1 and case 2 concerned diffusive mass transport, case 1 through a saturated soil and case 2 through a partially saturated soil. Case 1 is analogous to one-dimensional heat transport through a bar of constant cross section with depth, whereas case 2 is analogous to one-dimensional heat transport through a bar of increasing cross section with depth. This increasing cross-sectional area with depth is here referred to as the geometric influence.

To gain a better appreciation of the role played by the geometric influence on diffusive transport, contaminant transport solutions are again calculated for a soil liner with the moisture profile predicted from eq. [6], but this time having moisture content boundary conditions at the top and bottom of the liner. This approach enables the influence of a wide range of moisture profiles to be examined, and the geometric influence made apparent. It is noted that the moisture content at the bottom of the soil liner is taken to be the saturated moisture content in all cases.

Figure 16 shows three diagrams. On the horizontal axis is a normalized moisture content at the top of the soil liner. When the normalized moisture content equals one, there is a uniform moisture content equal to the saturated moisture content throughout the soil liner. The vertical axis shows the time in years for 1% (Fig. 16*a*), 50% (Fig. 16*b*), and 99% (Fig. 16*c*) of the initial mass of contaminant in the landfill to pass through the soil liner into the base aquifer.

The solid line in Fig. 16 represents the solution for the soil condition in case 1, that is, the time required for the specified percentage of contaminant mass to pass through a saturated soil liner. The line with long dashes represents the mass transfer times for various states of partial saturation determined for each of the moisture content boundary conditions. Comparing the solid line and the line with long dashes demonstrates the remarkable insensitivity of the time for 1%, 50%, and 99% of the contaminant mass to be transferred through the liner with respect to the moisture content at the top of the liner. A 50% reduction in moisture content at the top of the liner only increases the time for a specified percentage of contaminant mass transfer to pass through the liner by less than 10% in all three diagrams. To double the mass transfer time requires that the moisture content at the top of the liner be reduced to below 6% of the moisture content at the base.

To help confirm this extraordinary observation, a reexamination of the steady-state nondimensional solutions previously employed for transport modelling verification shows this behavior is also apparent, though it is not as extreme as that observed for transient conditions. For a diffusion-dominated transport process (i.e., Péclet number < 0.01), Fig. 4 shows that if the moisture content at the top of the layer is reduced to 50% of the moisture content at the bottom of the layer, the steady-state contaminant mass flux through the soil is reduced by only 30%, not 50% as might be expected intuitively. To reduce the contaminant mass transfer by 50% requires a 80% reduction in the moisture content at the top of the soil liner.

This mass transfer behavior for a diffusion-dominated process can be explained in the following way. As the volumetric moisture content is reduced, there is a reduction in the cross-sectional area available for mass transfer, thereby reducing the mass flux in proportion. However, the increase in moisture content with depth serves to dilute the contaminant concentration as it is transported through the soil liner. This dilution of contaminant with depth serves to increase the spatial concentration gradient at the top of the soil liner (seen in Fig. 3), and so in the case of a diffusion-dominated process, the rate of diffusive mass transfer is increased proportionally. These two effects, the reduction in cross-sectional area and the increased concentration gradient, oppose one another. The apparent insensitivity of contaminant mass transfer to the reduction in the moisture content at the top of the soil liner can be explained by the net effect of these two opposing influences.

On the other hand, the advective mass flux is constant if the Darcy velocity through the partially saturated soil liner is constant. This may be confirmed by examining the nondimensional solutions shown in Fig. 5. For a Péclet number of five, Fig. 5 shows that the contaminant mass flux through the liner is almost independent of the moisture content at the top of the soil layer. It is therefore apparent that a linearly increasing moisture content with depth does influence the rate of diffusive mass flux through an unsaturated soil liner, but it has no influence on the advective mass flux when a constant Darcy velocity is assumed.

Figures 11 and 12 indicate that taking the moisture dependence of the diffusion coefficient into account does have a very significant influence on the mass transfer characteristics of the liner. This is confirmed by reference to Fig. 16, the lines with short dashes indicating that, for each normalized moisture content at the top of the liner, the time for mass transfer through the liner is greatly increased. It is noted in passing that at some nonzero moisture content, the time for a percentage of the contaminant mass to pass through the liner becomes infinite. This is because the diffusion coefficient becomes zero at some nonzero moisture content (see eq. [12]).

It is further observed that, although diffusive mass transfer through a partially saturated liner is reduced as the moisture content is reduced, if the advective velocity through the liner is constant, the Péclet number for the soil liner increases, thereby setting a limit on the effectiveness of reductions in diffusive mass transfer as a means of controlling contaminant flux from the landfill. This indicates that control of the advective component of the contaminant transport should remain a central objective of liner design.

It is concluded that partial saturation of the soil liner does have a significant influence on diffusive contaminant mass transfer through a landfill liner, but the primary reason for the significant influence in this paper is not due to geometrical effects of partial saturation as may be commonly supposed, but is due to the moisture dependence of the diffusion coefficient. Although the geometrical effects of partial satu-

ration lead to a reduction in diffusive mass transfer through the soil liner, this investigation suggests that the moisture dependence of the diffusion coefficient is of primary importance.

As the diffusive mass transfer at low moisture content will be small, this suggests a role for partially saturated soil as a diffusive-transport barrier. Clearly though, a geomembrane may play a valuable role in controlling advection through the landfill liner, and therefore advective mass transport of the contaminant. It is apparent that each component of a geocomposite complements the other.

The main findings here suggest that an adequate characterization of the moisture dependence of the diffusion coefficient is essential for realistic modelling of diffusive mass transport through the unsaturated zone. However, it is clearly apparent that experimental difficulties involved in estimating the moisture dependence of the diffusion coefficient in clay soils pose a significant challenge for geotechnical experimentalists.

Conclusions

An approach for analyzing one-dimensional contaminant migration from a landfill through a partially saturated landfill liner has been described in detail. The quasi-linearized Richards equation was employed to predict the steady-state, unsaturated volumetric moisture distribution throughout a soil liner. It has been shown that for the example landfill with a 5 m thick soil liner, equilibrium moisture conditions were established over a time period that is comparatively small compared with the time required for contaminant mass transport through the liner. Therefore, employing a steady-state moisture distribution in the dispersion–advection equation is a reasonable approximation to make. A careful examination of the contaminant transport solutions for transport through an unsaturated soil liner suggests the following:

(1) Partial saturation of the soil liner leads to a reduction in cross-sectional area available for contaminant mass transport, which has the effect of decreasing the diffusive mass flux.

(2) The increase in moisture content with depth results in dilution of the contaminant with increasing depth in the soil liner. This has the effect of increasing the concentration gradient at the top of the soil liner and proportionally increasing the rate of diffusive mass transfer though the soil liner.

(3) The geometrical influences on contaminant transport through a liner described in (1) and (2) lead to two effects which oppose one another, thereby leading to a greatly reduced sensitivity of diffusive mass transport through a soil liner with respect to the moisture content at the top of the soil liner.

(4) Accounting for the dependence of the diffusion coefficient on the volumetric moisture content of the soil leads to a greatly reduced diffusive mass transfer through the soil liner. The very low diffusion coefficients for both fine- and coarse-grained soils at low volumetric moisture contents suggest a role for unsaturated soils as effective barriers to diffusive mass transport.

(5) Though experimentally difficult, it appears necessary to measure the moisture dependence of the diffusion coefficient for a realistic analysis of contaminant transport through a partially saturated soil liner.

(6) The analysis of the example geocomposite liner in this paper consisting of a geomembrane overlying a partially saturated soil liner suggests that, when advective transport is negligible, the relative beneficial effect of a geomembrane in containing the contaminant in a landfill is reduced as the degree of soil liner saturation decreases. In other words, at low moisture contents the primary barrier to diffusive mass transport will be the partially saturated soil. However, it is noted that the relative importance of advective transport through the liner will increase as the rate of diffusive mass transport through the liner decreases.

(7) The advective contaminant mass flux through the soil liner is not influenced by the degree of saturation if the Darcy velocity through the soil is constant.

For a properly functioning landfill liner the advective velocity is sufficiently small for diffusive mass transport to dominate advective mass transport. Therefore, conclusions (1)–(6) appear relevant for liner design with partially saturated soils. However, as noted in conclusion (7), the magnitude of the advective mass transport will set a lower limit on the effectiveness of a strategy of controlling diffusive mass transfer alone.

Although it is believed that general trends are revealed by the analysis of the example problem, every landfill design is unique and should be designed on an individual basis. Finally, it is noted that while it is likely that moisture dependence of the partitioning coefficient exerts a very important influence on contaminant mass transfer through a partially saturated soil liner, a detailed investigation of its significance is currently hampered by a dearth of experimental data.

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List of symbols

a_0, a_1,..., a_k: power series coefficients

A, B: empirically fitted constants in the expression to describe the spatial variability of ϑ

c^*: mapped concentration function
c: concentration of contaminant in the pore fluid
c_o: initial contaminant concentration
$c(s)$: transformed concentration function in terms of the transform parameters
c_{lo}: initial contaminant concentration at the top of the liner (i.e., within the landfill)
$c_l(t)$: contaminant concentration at the top of the liner (i.e., within the landfill)
$c_{T1}(t)$: contaminant concentration at the top of the soil liner
c_{Tn}, c_{Bn}: concentrations at the top and bottom, respectively, of the nth soil sublayer
$c_{BN}(t)$: contaminant concentration in the aquifer
$c_{BN,max}$: maximum contaminant concentration in the aquifer
D: effective hydrodynamic coefficient of the solute, usually assumed to be the sum of two components, namely, the diffusive component arising from the macroscopic description of mass transfer due to thermal agitation of the contaminant and the dispersive component arising from pore fluid velocity fluctuations about a mean value; D is also referred to as the molecular diffusion coefficient in the text
$D(\vartheta)$: function denoting the moisture dependence of the diffusion coefficient
D_f: free solution diffusion coefficient
D_g: membrane mass transfer coefficient
D_{ge}: diffusion coefficient of the contaminant in the geomembrane
D_m: moisture diffusivity in the unsaturated soil liner
D_{mo}: limiting moisture diffusivity as the moisture content approaches zero
D_{sat}: D for a saturated soil
E, G, J, L, X, Y: constants in the contaminant transport equation for an unsaturated soil; they are simple functions of the parameters A, B, Λ, Ω (and K_d if desired)
f_{N1}, f_{N2}: normalizing contaminant fluxes
$f_{BN}(c,\tau)$: contaminant flux into the aquifer from the soil liner above
f_{Tn}, f_{Bn}: contaminant fluxes at the top and bottom, respectively, of the nth soil sublayer
f_z: contaminant mass flux
h: thickness of a sublayer in the soil component of a geocomposite liner, so that $0 \le z \le h$
h_a: thickness of base aquifer
H: thickness of the soil component in a geocomposite liner
H_f: equivalent height of leachate; it is equal to the total volume of leachate at concentration c_{lo} divided by the plan area of the landfill
K_d: contaminant partitioning coefficient
L: landfill length
$m_{BN}(t)$: total mass of contaminant which has entered the aquifer from the base of the soil liner
m_{lo}: initial mass of contaminant in the landfill
n_b: effective porosity of the aquifer
n_c: effective porosity of the soil liner
N: total number of a soil sublayers in a multilayered soil liner profile
PN: Péclet number for the soil liner (= v_zH/n_cD)
r: rate of contaminant mass sink per unit volume of soil
s: Laplace transform parameter
t: time variable
t_{max}: time at which the maximum contaminant concentration occurs at the base of the liner
T_0, T_1,..., T_n: Chebyshev polynomials
v_b: advection in the base aquifer
v_t: average true linear velocity of the pore fluid in the z direction (positive downward)
v_z: Darcy moisture flow in the vertical direction (positive downward)
w_g: thickness of the geomembrane in the geocomposite liner
z^*: transformed vertical coordinate
z: vertical coordinate (positive downward)
α: soil parameter used in the estimation of the unsaturated permeability
γ: empirically fitted exponent
δ_j: vector of coefficients
θ: Kirchoff-transformed moisture content
Λ, Ω: empirically fitted constants
ρ_d: dry density of the soil
τ: tortuosity parameter
ϑ: volumetric moisture content of the soil
ϑ_*: reference value of ϑ
ϑ_l: moisture content at which the diffusion coefficient is zero valued
ϑ_T, ϑ_B: moisture contents at the top and bottom, respectively, of the soil liner
ϑ_{sat}: volumetric moisture content of the saturated soil; this is equal to the soil porosity
ς_j: vector of coefficients
Note: Overbars denote a Laplace-transformed quantity