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Solute transport under steady and transient conditions in biodegraded municipal solid waste

David Bendz
Department of Water Resources Engineering, Lund University, Lund, Sweden

Vijay P. Singh
Department of Civil and Environmental Engineering, Louisiana State University, Baton Rouge

Abstract. The transport of a conservative tracer (lithium) in a large (3.5 m^3) undisturbed municipal solid waste sample has been investigated under steady and fully transient conditions using a simple model. The model comprises a kinematic wave approximation for water movement, presented in a previous paper, and a strict convective solute flux law. The waste medium is conceptualized as a three-domain system consisting of a mobile domain (channels), an immobile fast domain, and an immobile slow domain. The mobile domain constitutes only a minor fraction of the medium, and the access to the major part of medium is constrained by diffusive transport. Thus the system is in a state of physical nonequilibrium. The fast immobile domain is the part of the matrix which surrounds the channels and forms the boundary between the channels and the matrix. Owing to its exposure to mobile water, which enhances the biodegradation process, this domain is assumed to be more porous and loose in its structure and therefore to respond faster to a change in solute concentration in the mobile domain compared to the regions deep inside the matrix. The diffusive mass exchange between the domains is modeled with two first-order mass transfer expressions coupled in series. Under transient conditions the system will also be in a state of hydraulic nonequilibrium. Hydraulic gradients build up between the channel domain and the matrix in response to the water input events. The gradients will govern a reversible flow and convective transport between the domains, here represented as a source/sink term in the governing equation. The model has been used to interpret and compare the results from a steady state experiment and an unsteady state experiment. By solely adjusting the size of the fraction of the immobile fast domain that is active in transferring solute, the model is capable of accurately reproducing the measured outflow breakthrough curves for both the steady and unsteady state experiments. During transient conditions the fraction of the immobile fast domain that is active in transferring solute is found to be about 65% larger than that under steady state conditions. It is therefore concluded that the water input pattern governs the size of the fraction of the immobile fast domain which, in turn, governs the solute residence time in the solid waste. It can be concluded that the contaminant transport process in landfills is likely to be in a state of both physical, hydraulic, and chemical nonequilibrium. The transport process for a conservative solute is here shown to be dominated by convective transport in the channels and a fast diffusive mass exchange with the surrounding matrix. This may imply that the observed leachate quality from landfills mainly reflects the biochemical conditions in these regions. The water input pattern is of great importance for the transport process since it governs the size of the fraction of the immobile fast domain which is active in transferring solute. This may be the reason for leachate quality to be seasonally or water flux dependent, which has been observed in several investigations. The result also has a significant practical implication for efforts to enhance the biodegradation process in landfills by recycling of the leachate.

1. Introduction

Our understanding of the conditions and the biochemical processes which govern the degradation rate of waste in landfills and emission fluxes from landfills is less than complete, and several researchers have emphasized the need for further research in the area of water flow and transport processes. *Straub and Lynch* [1982] identified water movement, leachate strength, and contaminant transport in the landfill interior as areas which require more attention. In modeling biodegradation and methane production, *Augenstein and Pacey* [1991], *Bogner and Spokas* [1993], and *El-Fadel et al.* [1996] stated that a most important research task, in order to improve the performance of degradation models, is to investigate the presence and mobility of water. *Zeiss and Major* [1993] and *Zeiss and Uguccioni* [1995, 1997] have pointed out the need to revise and develop the existing models for leachate production in order to

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better account for transient conditions and channel flow. In a comprehensive review of available models for flow and transport in landfills, *El-Fadel et al.* [1997b] stated that the transport processes in landfills is to a great extent an unknown field. The potential of the existing leachate quality models as prediction tools is very poor. Knowledge and data on spatial and temporal distribution of leachate quality and quantity, temperature, and microorganisms are missing. Future research within these areas is therefore in great demand.

The typical municipal solid waste landfill operates as a unmixed water deficient biological reactor, and the degradation process is accordingly inefficient [*Bogner and Spokas*, 1993; *El-Fadel*, 1996, 1997a]. Water constitutes the basic carrier for substances within a landfill. A high water content is therefore probably the most important prerequisite for anaerobic degradation processes [*Ehrig*, 1991; *Bogner and Spokas*, 1993]. The first step in the anaerobic degradation sequence, the hydrolysis, may be rate limiting for the whole degradation process [*Leuschner and Melden*, 1983]. This has also been indicated in an evaluation of microbial growth-gas production models by *El-Fadel et al.* [1996]. Second, a high water content facilitates the redistribution of nutrients and microorganisms within the landfill [*Augenstein and Pacey*, 1991; *Christensen and Kjeldsen*, 1989]. Also, the water flux has been shown to have a positive effect on the intensity of the biodegradation process [*Klink and Ham*, 1982]. The spatial variation of water content is large and varies from saturated conditions to complete dryness. Acknowledging this, leachate recirculation has became the most common and efficient method to enhance the biodegradation process [*Yuen et al.*, 1995]. In addition to optimizing the environmental conditions for the anaerobic process, the method also has the potential to reduce the level of contaminants in the leachate because of anaerobic treatment in the landfill interior and dilution. A large amount of study and research has been conducted both in laboratory and pilot scale [*Barlaz et al.*, 1987; *Pohland*, 1975; *Stegmann and Spendlin*, 1989] and full scale [*Barber and Maris*, 1984; 1992; *Pacey*, 1989; *Reinhart*, 1996; *Reinhart and Al-Yousfi*, 1996; *Stegmann and Spendlin*, 1986, 1989].

In reviews by *Bendz et al.* [1997] and *El-Fadel et al.* [1997b] it is found that the prevailing approach for modeling of transport in a solid waste medium is to apply the Richards equation to determine the space-time history of the water velocity and to incorporate this into the convection-dispersion equation (CDE) to give the solute concentration [*Ahmed et al.*, 1992; *Demetracopoulus et al.*, 1986; *Korfiatis et al.*, 1984; *Lee et al.*, 1991; *Straub and Lynch*, 1982; *Vincent et al.*, 1991]. This approach can be questioned since the basic assumption of the CDE, which justifies the formulation of the hydrodynamic dispersion flux as a diffusive flux, may not be fulfilled because of the considerable heterogeneity of the waste medium [*Bendz et al.*, 1998]. Further, the presence of fast channel flow, which has been observed in several investigations [*Bengtsson et al.*, 1994; *Blakey*, 1982; *Blight et al.*, 1992; *Burrows et al.*, 1997; *Harris*, 1979; *Ham and Bookter*, 1982; *Holmes*, 1983; *Korfiates et al.*, 1984; *Walsh and Kinman*, 1979; *Zeiss and Major*, 1993; *Zeiss and Uguccioni*, 1995, 1997], constitutes a deviation from Richards' assumption of a perfect correlation between capillary diffusivity and capillary potential [*Germann*, 1990]. No models for transport processes in landfills are known to us where the nonideal transport process, resulting from the heterogeneity of the medium and the nonuniform flow field, have been taken into account.

The transport of contaminants in heterogeneous media, such as waste in landfills, is characteristically a nonideal process and is in a state of physical, chemical, and hydraulic nonequilibrium. Chemical nonequilibrium is associated with sorption nonequilibrium and intrasorbent diffusion, whereas physical and hydraulic nonequilibrium is due to a nonuniform flow field which, in turn, is a result of physical heterogeneity of the medium. The system is said to be in physical equilibrium when differences in solute mobility cause concentration gradients between the regions and access to certain parts of the medium may be restricted to diffusive transport.

Systems which are in a state of nonequilibrium are commonly conceptualized as composed of two or several domains based on the physical or chemical heterogeneity. As shown in the reviews by *Brusseau and Rao* [1990], *Gerke and van Genuchten* [1993], and *Sardin et al.* [1991], the multidomain concept has been widely used to describe physical nonequilibrium when modeling flow and transport processes in soil and rocks exhibiting various kinds of heterogeneities, such as macropores, fractures, and fissures. In all of these cases, significant deviations from a uniform flow field, because of fast flow in restricted pathways, have been observed. The mobile-immobile (MIM) concept describing physical nonequilibrium by *Deans* [1963] and *Coats and Smith* [1964] and further developed by *van Genuchten and Wierenga* [1976] to include sorption is by far the most well-known two-domain model and has been employed in numerous studies. The diffusive mass transfer between the regions is assumed to be governed by the concentration difference and a mass transfer coefficient. The MIM concept is useful in describing the effects of stagnant regions such as the tailing phenomenon. However, when used as a simulation model, it has a certain weakness since its parameters cannot be measured independently but must be determined by simultaneously fitting the parameters to the outflow breakthrough curve (BTC) [*Jury et al.*, 1991; *White*, 1985]. *Brusseau et al.* [1994] lent the dual-domain model by *van Genuchten and Wierenga* [1976] further credibility by conducting tracer experiments in well-characterized soil and comparing model parameters obtained by curve fitting to the experimental data with parameters determined independently. Good agreement suggested that the model parameters determined by curve fitting represent actual physical phenomena.

Although the mass transfer coefficient is commonly regarded as a constant, it is likely that both convective [*Gwo et al.*, 1996] and diffusive mass transfer coefficients [*Gwo et al.*, 1996; *Reedy et al.*, 1996] are time dependent. In a theoretical analysis by *Rao et al.* [1980] an explicit expression was derived, relating the mass transfer coefficient for porous spherical aggregates to the physical properties of the system. It was shown that the mass transfer coefficient is dependent on the effective molecular diffusion coefficient, the water content inside and outside the aggregates, sphere radius, and the time available for diffusion. The dependence on the latter variable implies that the mass transfer coefficient is positively related to the pore water velocity [*Nkedi-Kizza et al.*, 1983; *Reedy et al.*, 1996]. *Haggerty and Gorelick* [1995] presented a multirate model that uses a set of different first-order coefficients. It is demonstrated that a mass transfer model with a distribution of first-order mass transfer coefficients is mathematically equivalent to the physical diffusive transfer model. By employing a certain probability distribution for the mass transfer coefficient, specific diffusion models, such as the spherical, cylindrical, and layered models, can be obtained. A multirate formulation can

therefore be seen as a reduced form of the physical diffusive model.

Additional complexity associated with the impact of preferential flow on the transport process is introduced under transient conditions. Important factors that govern the impact are initial and boundary conditions and hydraulic properties of the medium such as particle size and spatial pattern of the preferential flow paths. Also, the influence of physical properties of the medium will not be constant during transient conditions. For example, the fraction of mobile water has been shown to be dependent on the flux [*Nkedi-Kizza et al.*, 1983] and will not remain constant under transient conditions [*Brusseau and Rao*, 1990]. Under transient conditions, hydraulic gradients develop between regions of fast flow and regions of more or less stagnant water volumes. This results in water flow and convective transport between the regions, driven by the hydraulic gradient, which strives to reestablish hydraulic equilibrium [*Reedy et al.*, 1996; *Hutson and Wagenet*, 1995]. If the flow becomes steady, the hydraulic gradients will attenuate and equilibrium will be established. In response to an unsteady water input pattern at the boundary the flow system will therefore go through perturbation and equilibrium cycles [*Gwo et al.*, 1996]. The magnitude of these cycles will lessen with depth. Under transient conditions the mass transfer flux consists of a component driven by the concentration difference and one convective component. *Gwo et al.* [1996] employed a one-dimensional three-domain model and studied the coupled effects of convective and diffusive mass transfer under transient conditions in a structured soil. It was found that convective transport could counteract diffusive mass transfer under certain conditions. Further, it was shown that for large mass exchange rates the diffusive mass transfer gave a significantly larger contribution to the mass transport between the domains compared to the convective mass transfer, which was attributed to a fast diminishing of the hydraulic gradients. For smaller exchange rates, diffusive and convective mass transfer were found to be comparable.

The extent of physical nonequilibrium can be estimated by stopping the steady state flow process for a period of time so that hydraulic equilibrium could be established; the diffusive mass transfer between the regions is then the only process which is active. This is the flow interruption technique of *Brusseau et al.* [1989, 1997] which can help to identify the presence of various rate-limiting processes for a system in nonequilibrium, such as rate-limited sorption, diffusive mass transfer, and transformation reactions. *Brusseau et al.* [1997] demonstrated the usefulness of the method by performing experiments using a selection of setups and investigating specific process pairs such as physical nonequilibrium versus heterogeneity. Both phenomena cause spreading of the breakthrough curves, but only physical nonequilibrium causes a significant perturbation of the breakthrough curve. For systems influenced by rate-limited sorption or physical nonequilibrium, the concentration drops if the flow is interrupted during the rising part of the BTC and increases if the interruption is made during the declining phase.

We have previously applied a kinematic wave model to model the infiltration and drainage of a sequence of wetting events in a large-scale undisturbed municipal solid waste sample [*Bendz et al.*, 1998]. Here the flow model was coupled to a piston flux law for solute transport to produce the space and time history of solute concentration. The MIM conceptualization of the medium as proposed by *van Genuchten and Wierenga* [1976] was employed. The MIM model was further discretized by dividing the immobile domain into two subdomains. The model was used here to parameterize and interpret results by calibrating it with data from solute experiments performed under steady and unsteady conditions. The experimental setup and the unsteady flow conditions are identical to those described by *Bendz et al.* [1998].

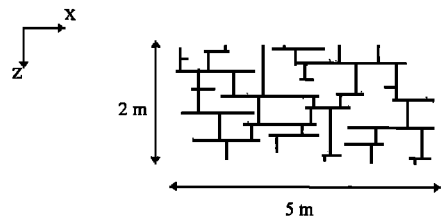

Figure 1. Schematic illustration of the flow paths in the landfill interior [*Bendz et al.*, 1997].

The objective of this study was to investigate the nonideal behavior of the solute transport process in solid waste by calibrating the model to breakthrough curves from experiments performed under steady and unsteady conditions. A conservative solute was used, and thus this study is restricted to physical and hydraulic nonequilibrium mechanisms.

2. Theory

2.1. Conceptual Model

Waste in landfills is a highly heterogeneous media both in terms of geometry and composition. Owing to the disposal and compaction procedure a strong horizontal stratification can be observed which makes the waste medium anisotropic in the vertical plane. Although heterogeneity and anisotropy in landfills have been observed by several researchers [e.g., *Burrows*, 1997; *Oweis et al.*, 1990], a quantitative description of their spatial variability is still lacking. We have previously suggested [*Bendz et al.*, 1997] that the landfill waste medium can be conceptualized physically as discretely hierarchical, being composed of porous lens-shaped elements with partially impermeable surfaces (compacted and partially torn refuse bags). The lenses are separated by a network of voids or channels. The domains, in which flow and transport take place, can be easily distinguished as the interior of the lenses and the structural voids which form channels. Regarding the internal geometry of the landfill, the channel flow pattern can be described as follows: Owing to stratification and the presence of impermeable materials such as plastic a significant portion of the flow takes place in the horizontal direction [*Burrows*, 1997; *Knox*, 1996]. These flow paths at different levels are connected by vertical shortcuts. This leads to a network of flow paths, similar to those in fractured rocks or fissured media. A schematic illustration of this flow pattern in two dimensions is given in Figure 1.

On the basis of this we suggested that the landfilled waste medium can be discretized into a matrix domain and a channel domain, which would allow separate assumptions regarding the flow and transport processes to be made in each domain [*Bendz et al.*, 1997]. The channel domain was assumed to consist of a network of flow paths in which the movement of water is dominated by gravity, whereas in the matrix domain the capillary forces are dominant. The matrix domain is composed of the solid phase, a region in which water can be held against

gravity by capillary forces, and a drainable water region. The space which is not filled with water is occupied by air or biogas. The water in the capillary region of the matrix domain is, in comparison with the fast channel flow, regarded to be stagnant and will hereafter be referred to as immobile. The water in the channels will be referred to as mobile. The maximum water content of the capillary region is identical to the field capacity. The vertical flow of water in the drainable region of the matrix is assumed to be negligible. Water in this region is assumed to move mainly in the horizontal direction until it intersects the channel domain where gravity dominates and the water rapidly flows downward. This assumption can be justified by an apparent horizontal stratification in combination with impermeable materials of landfill waste. There may be a reversible water exchange, governed by a hydraulic gradient, between the channel domain and the drainable region of the matrix domain that surrounds the channels.

Solute transport in the channels is assumed to be strictly convective and is described by a simple piston flux law. Although a solute can move in the matrix domain by diffusion, this form of transport is assumed to be negligible in comparison with the fast convective transport in the channel domain. Solute transport between the regions is due to convective and diffusive mass transfer. Since the channel domain is very small, the access to a large part of the medium is constrained by diffusive transport, and the system is therefore usually in a state of physical nonequilibrium. Under transient conditions, hydraulic gradients build up between the channel domain and the matrix in response to the water input events. The system will now be in a state of both physical and hydraulic nonequilibrium, and the gradients will govern a reversible flow and convective transport between the domains. This convective mass transfer is represented as a source/sink term in the governing equation.

The immobile domain is divided into two subdomains since it is likely that the part of the matrix which surrounds the channels and forms the boundary between the channels and the matrix is more porous and loose because of its exposure to mobile water which enhances the biodegradation process. This region will therefore respond faster to the concentration in the mobile domain compared to the regions deep inside the matrix. In analogy with the dynamic soil region of instantaneous sorption in the MIM model we assume here that the diffusive mass transfer between the mobile domain and a fraction of the immobile region, the fast immobile domain, is fast enough so that the two regions have had enough time to mix completely before the mobile water reaches the lower boundary of the column. While for the rest of the immobile region, the transport of solute is diffusion limited. This defines the slow immobile domain. The solute that diffuses into the deeper regions of the matrix is not so easily recovered on a short time basis. The size of the mobile and total immobile domains is defined by their physical characteristics. The fraction of the immobile domain that is active depends on the size of the effective area which forms the boundary between the mobile and the immobile parts of the system. The size of this area is governed by the physical properties of the medium, but the accessibility to the fast immobile domain is dependent on the flow conditions. Both a high water flux and hydraulic gradients resulting in lateral flow, which will develop under transient conditions, will increase the accessibility to the fast immobile domain. *Jasper et al.* [1985] suggested that additional flow routes may be developed during periods of high infiltration rate, and the flow pattern may therefore be dependent on the boundary conditions. *Akesson and Nilsson* [1997] investigated the seasonal variability of leachate production and quality from two pilot-scale landfills. They found that a number of leachate quality parameters were correlated to the leachate flux such that the biochemical conditions in the landfill interior appeared to alternate between methanogenic at low flux rates and acidogenic at high flux rates. Several hypothetical configurations of spatially variable biochemical conditions were proposed and evaluated. One of the configurations that could not be rejected was a pore-scale configuration where an isolated pore domain, containing stagnant water held by capillary forces, was connected to a mobile water system during periods of high flux.

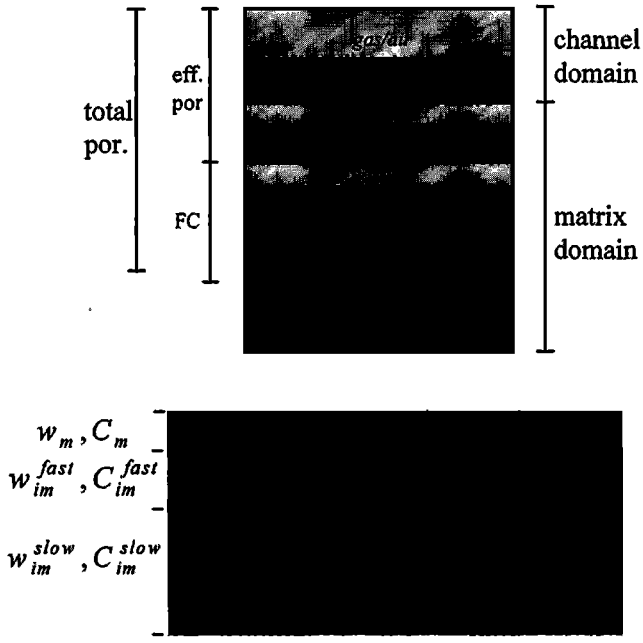

Figure 2. Illustration of the different domains. The w denotes the water content as a volumetric fraction of the total volume; the subscripts m and im denote the fractions of mobile and immobile water regions, respectively; the superscripts fast and slow denote the fast and slow parts of the immobile domain, respectively; FC is the field capacity; and w_{mat} is the drainable water content in the matrix. Note that the total porosity is assumed to be equal to the sum of the effective porosity and the field capacity. The noninterconnected closed pore volume is neglected. The diffusive exchange between the domains is modeled with two first-order mass transfer expressions coupled in series.

The diffusive transport is modeled with two first-order mass transfer expressions coupled in series. The domains and regions described above are illustrated in Figure 2.

2.2. Flow Model

The space and time history of water content in the channel domain is given by the kinematic wave model presented in our earlier paper [*Bendz et al.*, 1998].

The framework of the flow model is summarized below. The flow in the channel domain is characterized as a thin viscous film with free surface. The water film moves, under the influence of gravity, along the surfaces that constitute the boundaries of the channels. On the basis of this the following two-parameter power function, originally proposed by *Beven and*

Germann [1981] for macropore flow in soils, was employed as a macroscopic flux law for channel flow in landfill waste.

$$q = bw^a \tag{1}$$

where q is the water flux density (m s^{-1}) in the macropores per unit cross-sectional area of soil, a is a dimensionless exponent, and b is the macropore conductance (m s^{-1}) which can be interpreted as the integrated effect of surface, geometrical, and spatial characteristics of the flow path. The amount of water in the macropores is expressed as a volume fraction of the soil w rather than the thickness of a water film.

We determined the parameters in the flux law, for the experimental setup that is used here, by a calibration procedure and found them to be [*Bendz et al.*, 1998]:

$$\begin{cases} a = 3.05 \\ b = 5.24 \text{ m s}^{-1} \end{cases}$$

The exponent a was reasoned by *Germann and Di Pietro* [1996] to be dependent on the internal geometry of the medium, the initial water content, and the boundary conditions affecting the water content at the surface. The fluid-mechanical interpretation of the parameter a was summarized as a measure of the impact of the stagnant parts of a flow system on the mobile parts. For macropore flow the parameter a was expected to be in the range of $2 \leq a < \approx 8$, where $a = 2$ indicates laminar flow in a cylindrical pipe (which may occur if the medium is saturated) and $a = 3$ indicates flow in planar cracks. Turbulent flow is indicated when $a < 2$. Higher values of a indicate increasingly tortuous flow. Completely dispersive flow corresponds to $a > \approx 10$. According to this fluid-mechanical interpretation of the magnitude of a it was concluded that the channel flow in the waste medium could be characterized as flow along a planar surface, that is, flow along the boundaries of the channels.

The governing equations in the flow model are the law of conservation of mass and the flux law (1). The conservation of mass equation can be expressed as

$$\frac{\partial w_m}{\partial t} + \frac{\partial q}{\partial z} = -S \tag{2}$$

where the subscript m denotes mobile and corresponds to the channel flow, S is the rate of water exchange between the channel domain and the matrix domain (per second), t is the time (seconds), and z is the depth (meters).

Inserting (1) into (2) and, for a kinematic wave treatment, assuming that at a fixed z, q is a function of w alone yields a kinematic wave equation,

$$\frac{\partial w_m}{\partial t} + c\frac{\partial w_m}{\partial z} = -S \tag{3}$$

in which

$$c = \frac{\partial q}{\partial w_m} = abw_m^{a-1} \tag{4}$$

where c is the wave celerity (m s^{-1}). The last term follows from (1) and inserted into (3) gives

$$\frac{dw_m}{\partial t} + abw_m^{a-1}\frac{\partial w_m}{\partial z} = -S \tag{5}$$

which can be solved analytically by the method of characteristics. Equation (5) is then reduced to the following system of characteristic equations:

$$\frac{dw_m}{dt} = -S \tag{6}$$

$$\frac{dz}{dt} = abw_m^{a-1} \tag{7}$$

For a square pulse input q_u at the surface, starting at $t = 0$ and ending at $t = T$, the following initial and boundary conditions can be assumed:

$$w_m(z, 0) = f(z) \tag{8}$$

$$w_m(0, t_s) = \begin{cases} 0 & t_s \leq 0,\ t_s \geq T \\ w_u & 0 < t_s < T \end{cases} \tag{9}$$

where $f(z)$ is an arbitrary function describing the initial water content, w_u is the water content in the channels at the upper boundary, $z = 0$ during the square pulse input, and t_S is the point on the time axis where the characteristic defined by (7) starts. Given the initial and boundary conditions, (6) and (7) can be solved. The solution of (7) gives the time and space history of the characteristic along which the water content defined by the solution of (6) travels. The solution is given by *Bendz et al.* [1998] and will not be described here.

For a water input event at the surface the water content will increase abruptly and cause a discontinuity in the water content profile. This discontinuity will move downward as a shock front maintaining a sharp interface between the water content of the front and the water content ahead. By obeying the continuity equation, the shock velocity in the case where w decreases with depth can be written as [*Smith*, 1983]:

$$\frac{dz}{dt} = \frac{q(w) - q(w^*)}{w - w^*} \tag{10}$$

where w is the water content, defined by (6), that propagates down along its characteristic defined by (7) and w^* is the water content in the waste medium ahead of the wetting front.

2.3. Transport Model

The continuity equation for solute is written as

$$\frac{\partial wC}{\partial t} + \frac{\partial q_s}{\partial z} = -SC_m \tag{11}$$

where C is the total solute concentration (g m^{-3}), w is the total water content expressed as a fraction of the total volume, q_s is the solute flux (g (m^2 s)$^{-1}$) and SC_m represents the loss of solute due to convection into adjacent voids and pores in the matrix. Since the solute is assumed to be conservative, adsorption is not taken into account. Inspired by *van Genuchten and Wierenga* [1976], the total concentration is written as

$$wC = w_mC_m + fw_{im}^{\text{fast}}C_{im}^{\text{fast}} + w_{im}^{\text{slow}}C_{im}^{\text{slow}} \tag{12}$$

As pointed out in section 2.1, the accessibility to the fast immobile domain is dependent on the flow conditions. In (12), f denotes the fraction of the fast immobile domain that is active in transferring solute. The immobile concentration C_{im}^{fast} is associated with the active part of the fast immobile domain. The immobile concentrations C_{im}^{fast} and C_{im}^{slow} are treated as constant in space so that there are no concentration gradients within the immobile domains. The diffusional mass transfer between the fast immobile domain and the slow immobile domain is described by a first-order expression:

$$w_{im}^{slow} \frac{\partial C_{im}^{slow}}{\partial t} = \beta(C_{im}^{fast} - C_{im}^{slow}) \quad (13)$$

Since the two immobile domains are coupled in series, the solute concentration in the fast immobile domain is defined by

$$fw_{im}^{fast} \frac{\partial C_{im}^{fast}}{\partial t} = \alpha(C_m - C_{im}^{fast}) - \beta(C_{im}^{fast} - C_{im}^{slow}) \quad (14)$$

where α and β are the mass transfer rate coefficients (per second) for transport between the mobile–fast immobile domains and the fast immobile–slow immobile domains, respectively. The transfer rate coefficients can be interpreted as a diffusion coefficient divided by an average diffusional path length.

The mass flux law of the commonly used CDE is

$$q_s = q_w C_m - D \frac{\partial C_m}{\partial z} \quad (15)$$

where q_w is the water flux (m s^{-1}) and D is the lumped dispersion-diffusion coefficient. However, since the basic assumption of the CDE could not be justified because of the heterogeneity of the medium and for simplicity, the dissipative term is neglected and the concentration front is assumed to move as a piston. The solute flux law becomes

$$q_s = q_w C_m \quad (16)$$

The observed tailing phenomenon is represented in the model solely by the mass transfer between the mobile and immobile regions.

Combining (11), (12), and (16) and assuming that the immobile water content is constant yields

$$w_m \frac{\partial C_m}{dt} + C_m \frac{\partial w_m}{\partial t} + fw_{im}^{fast} \frac{\partial C_{im}^{fast}}{\partial t} + w_{im}^{slow} \frac{\partial C_{im}^{slow}}{\partial t} + q_w \frac{\partial C_m}{\partial z} + C_m \frac{\partial q_w}{\partial z} = -SC_m \quad (17)$$

Noting that the sum of the second term and the sixth term on the left-hand side equals the term on the right-hand side, (17) becomes

$$w_m \frac{\partial C_m}{\partial t} + fw_{im}^{fast} \frac{\partial C_{im}^{fast}}{\partial t} + w_{im}^{slow} \frac{\partial C_{im}^{slow}}{\partial t} + q_w \frac{\partial C_m}{\partial z} = 0 \quad (18)$$

and inserting (13) and (14) into (18) gives

$$\frac{\partial C_m}{\partial t} + \frac{q_w}{w_m} \frac{\partial C_m}{\partial z} = -\frac{\alpha}{w_m}(C_m - C_{im}^{fast}) \quad (19)$$

Equation (19) is a linear first-order partial differential equation and can be solved using the method of characteristics. The characteristic equations are

$$\frac{dC_m}{dt} = -\frac{\alpha}{w_m}(C_m - C_{im}^{fast}) \quad (20)$$

$$\frac{dz}{dt} = \frac{q_w}{w_m} = u \quad (21)$$

According to (21) the concentration celerity is the same as the average water flow velocity. Thus the solute will travel slower than the wave celerity defined by (4).

The initial and boundary conditions are

$$C_m(0, t) = C_u \qquad 0 \le t \le T_p \quad (22a)$$

$$C_m(0, t) = 0 \qquad t < 0 \text{ and } t > T_p \quad (22b)$$

$$C_{im}^{fast}(z, 0) = C_{im}^{slow}(z, 0) = 0 \qquad 0 \le z \le Z \quad (22c)$$

where C_u is the solute concentration in the applied pulse and $0 \le t \le T_p$ is the time segment during which solute is applied at the surface.

2.4. Solutions

When the immobile concentration and the mobile water content are assumed to be constant in time, (20), under the boundary condition given by (22a), has the following solution:

$$C_m(t, t_s) = C_{im}^{fast} + (C_u - C_{im}^{fast}) e^{-\frac{\alpha}{w_m}(t - t_s)} \qquad 0 \le t_s \le T_p \quad (23)$$

The solution shows that C_m declines exponentially along the characteristic defined by (21). The water content is constant only during steady state flow, and the immobile concentration in the fast region shows large temporal fluctuations and may therefore not be regarded as constant. Accordingly, (23) is only applicable for short time intervals Δt and can then be written

$$C_m(t + \Delta t, t_s) = C(t)_{im}^{fast} + (C_m(t, t_s) - C(t)_{im}^{fast}) e^{-\frac{\alpha}{w_m(z,t)}\Delta t} \qquad 0 \le t_s \le T_p \quad (24)$$

The mobile water content at space-time coordinates (z, t) and $w_m(z, t)$ is given by the flow model of *Bendz et al.* [1998].

Substitution of (14) into (20) gives

$$\frac{dC_m}{dt} = -\frac{1}{w_m}\left[fw_{im}^{fast} \frac{\partial C_{im}^{fast}}{\partial t} + \beta(C_{im}^{fast} - C_{im}^{slow})\right] \quad (25)$$

The immobile concentration in the fast region at time $t + \Delta t$ is calculated by employing a simple numerical scheme and solving (25) for $C_{im}^{fast}(t + \Delta t)$:

$$C_{im}^{fast}(t + \Delta t) = C_{im}^{fast}(t) - \frac{1}{fw_{im}^{fast}}\left\{\beta[C_{im}^{fast}(t) - C_{im}^{slow}(t)]\Delta t + \frac{1}{Z}\sum_{t_s = t_s^{min}}^{t_s = t_s^{max}} \bar{w}_m \Delta z [C_m(t + \Delta t, t_s) - C_m(t, t_s)]\right\} \quad (26)$$

where

$$\Delta z = \frac{1}{4}[z(t + \Delta t, t_s - 1) - z(t + \Delta t, t_s + 1) + z(t, t_s - 1) - z(t, t_s + 1)]$$

$$\bar{w}_m = \frac{1}{2}[w_m(t + \Delta t, t_s) + w_m(t, t_s)]$$

t_s^{min} and t_s^{max} denote the time of origin of the "oldest" and the "newest" characteristics in the ensemble, respectively, and Z is the total depth of the sample.

The BTC at the lower boundary Z can be determined by a routing procedure, using (24) and (26) under the appropriate initial and boundary conditions. For each time step Δt and under the condition that there is water input P, a characteristic is added such that $t_s^{max} = t + \Delta t$, and it carries the boundary conditions given by (22a) and (22b). The mobile solute con-

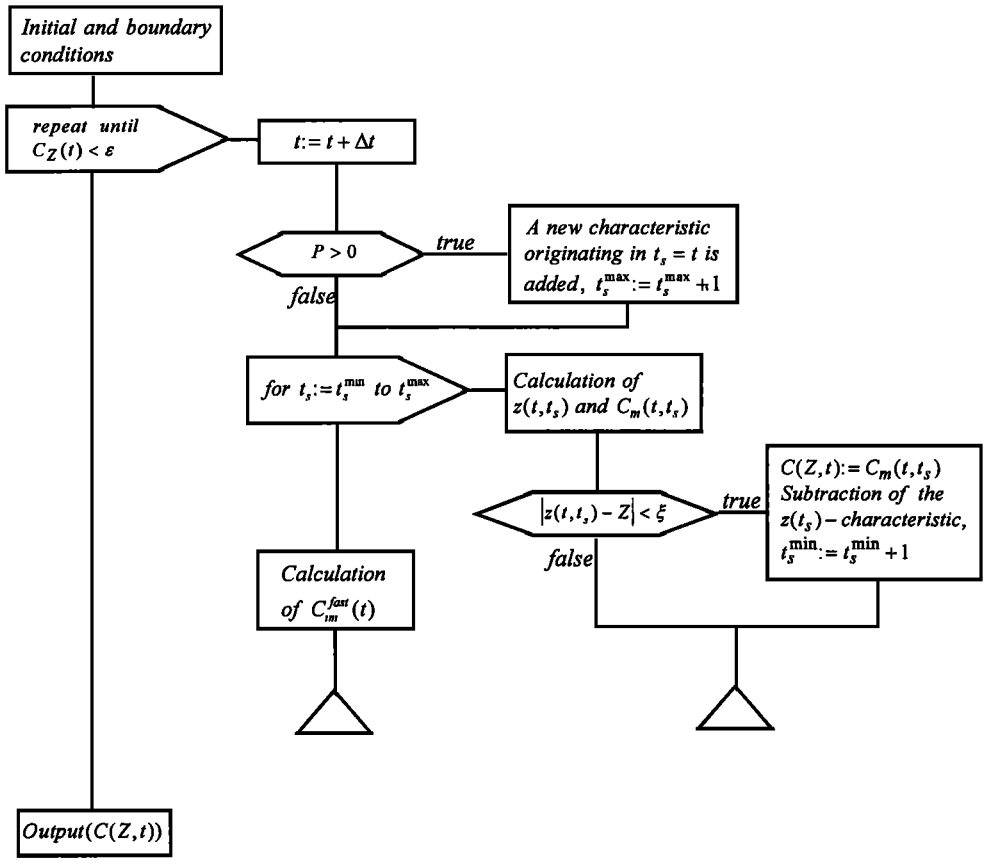

Figure 3. The routing procedure presented as a flow chart.

centration is calculated with equation (24) for the whole ensemble of solute characteristics for each time step. By inserting expressions for $w(z, t)$, given by *Bendz et al.* [1998], into (21) and employing a Runga-Kutta procedure, the z coordinate at the new time, $z(t + \Delta t, t_s)$, can be calculated for each solute characteristic. If a characteristic reaches the lower boundary, defined by $|z(t + \Delta t, t_s) - Z| < \xi$, where ξ denotes an arbitrary small number, $C_m(t + \Delta t, t_s)$ is registered as the outflow concentration, $C(Z, t + \Delta t)$. The characteristic is thereafter removed from the ensemble of characteristics so that the time of origin of the "oldest" characteristic t_s^{min} becomes $t_s^{min} + \Delta t$. The new immobile solute concentration is then calculated with (26). A schematic illustration of the procedure is given in Figure 3.

3. Materials and Methods

3.1. Setup

Rosqvist and Bendz [1999] have described in detail the sampling procedure, experimental setup, and the total number of experiments performed on the column. Therefore only the part of the experimental setup that is of direct interest in this study is described here.

The sample was taken in 1995 from a 22-year-old test cell, originally containing shredded household waste. The dimensions of the sample are 1.93 m in diameter and 1.20 m in height. The composition of the waste at the time of disposal is given in Table 1. The test cell was compacted with conventional methods to a dry density of 380 kg m^{-3}.

Since the late 1970s the cell has been laying under the groundwater table. In a recent investigation of the current composition and geometrical forms of the waste, *Flyhammar et al.* [1998] found that the fraction of paper had decreased by more than 40% by weight and that the easily degradable materials were almost completely degraded.

The current properties of the sample are summarized in Table 2 [*Bendz et al.*, 1998]. The total porosity was determined by saturating the sample; that is, the presence of closed air-filled voids was neglected.

According to our observations the waste was highly compacted, stratified, and tightly clustered. The increase in the dry density, compared to the original value, is an effect of biodegradation and settlement. The sample may represent highly compacted well-degraded waste, which can be expected to be found at larger depths in landfills.

With the aid of an excavator the sample was taken by a

Table 1. Original Composition of the Waste in the Test Cell

Material	Composition, wt %
Sludge	35
Paper, Biodegradables, glass	47
Textiles	6
Metal	6
Plastic	3
Wood, timber	3

Source is *Persson and Rylander* [1977].

Table 2. Measured Properties for the Column Sample

Property	Value
Height, m	1.20
Diameter, m	1.93
Dry density, kg m^{-3}	590
Porosity, m^3 m^{-3}	0.53
Effective porosity, m^3 m^{-3}	0.12
Field capacity, m^3 m^{-3}	0.41

Source is *Bendz et al.* [1998].

punching technique. A steel cylinder, measuring 1.93 m in diameter, was pressed down into the waste. In this way a cylindrical volume was cut out. At the depth of 1.2 m a steel sheet was forced in under the cylinder and fastened in place. The cylinder was then lifted up and brought to the laboratory where the steel sheet was removed and the cylinder was installed on a stand equipped with a drainage layer and a drainage pipe. On top of the cylinder an irrigation system of 19 microsprinklers was installed on a bar that was kept rotating to insure that the sample was irrigated evenly. The irrigation flux was measured with an electronic flowmeter, and the outflow flux was measured with a tipping bucket device connected to a data logger. The column sample was at field capacity at the beginning of the experiments.

3.2. Experiments

Two solute experiments, under steady and unsteady conditions, are presented in this paper. Lithium bromide (LiBr) was used as a conservative tracer in both experiments. The same mass of LiBr was used in both experiments.

The first solute experiment was performed during steady unsaturated flow to calibrate the model parameters. The flow was kept steady at 2.87×10^{-5} m^3 s^{-1}, corresponding to a flux density of 9.80×10^{-6} m s^{-1}. Irrigation was then shut off, and a 6-min solute pulse with a concentration of 428 mg L^{-1} (Li^+) was applied on the surface before irrigation was reestablished.

The second solute experiment was conducted under unsteady conditions. The unsteady state experiment was performed by applying a periodic square pulse with an average flow equal to the flow in the calibration experiment. Each pulse was 6 min long with a water flux of 1.14×10^{-4} m^3 s^{-1}, corresponding to a water flux density of 3.92×10^{-5} m s^{-1}. The pulses were applied with a 24-min period. During the first 90 s of the first pulse, water with a lithium concentration of 405 mg L^{-1} (Li^+) was added. Pure water was used during the remaining 270 s of the pulse. Water samples were collected at the column outflow every minute up to 40 min after the solute was applied, every 5 min from 40 to 100 min, every 25 min from 100 to 200 minutes, and, finally, every 50 min. The last sample was taken at 500 min for the steady state experiment and at 250 min for the unsteady experiment. The samples were analyzed for concentration of Li^+ using an atomic photospectrometer. The results of the solute experiments are shown in Figure 4.

The observed breakthrough curves show early steep rises and long tails, which support the hypothesis of fast macropore flow in a small fraction of the total porosity and a large stagnant water volume from which the solute slowly diffuses out into the mobile part of the system. This is further supported by the breakthrough curve in the unsteady state experiment which shows a significant response to the water input events. The water pulses cause a decrease in concentration due to dilution, whereas the concentration increases during the periods of drainage, which is expected for a system in a state of physical nonequilibrium.

The tailing is more pronounced for the unsteady state experiment in which there is temporary storage of water due to high water flux during the pulses, which force water into voids and dead-end pores in the drainable region of the matrix. This causes the solute to diffuse deeper into the immobile region where it is not so easily recovered. This finding is supported by *Hutson and Wagenet* [1995], who conducted simulations with a multidomain model and found that if solute is introduced into the immobile domain of a structured soil by convection it may be quickly absorbed and retained for a long time if the transport back to the mobile domains is by diffusive mass transfer only. In this case the water flux from the channel domain into the matrix during the pulses is much larger than the water flux from the matrix back to the channel domain [*Bendz et al.*, 1998]. The finding by *Hutson and Wagenet* [1995] is therefore to some extent applicable here.

The breakthrough time in the steady state experiment, defined as the time at which the breakthrough curve reaches its peak, corresponds to a volume of about 0.046 m^3 which has flowed through the system, which represents about 1.3% of the total volume or 11% of the effective porosity. In the unsteady state experiment every water pulse input corresponds to a peak in the BTC. The first two peaks are of equal magnitude and correspond to water displacement volumes of 0.043 m^3 and 0.082 m^3, respectively. If the breakthrough time is taken as the average of the first two peaks, the water displacement volume represents about 1.7% of the total volume and 14% of the effective porosity. There is apparently no significant difference between the two experiments in terms of breakthrough time. However, by fitting a log-normal distribution function to the experimental data, *Rosqvist and Bendz* [1999] showed that the expected residence time for a solute particle is twice as long in the unsteady state experiment as in the steady state experi-

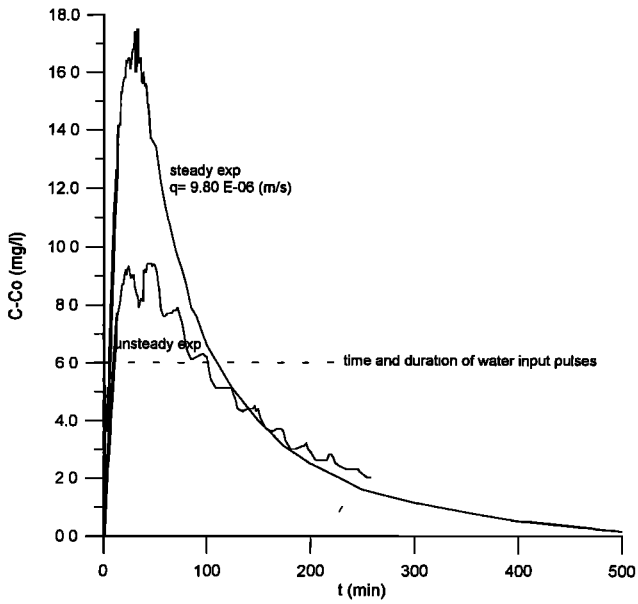

Figure 4. Breakthrough curves (BTCs) for steady and unsteady state solute experiments, registered at the outflow, of the column (Z = 1.2 m). The time and duration of the applied water pulses are marked by bold dashes.

ment. The expected residence time determined corresponded to water displacements of 6.1% and 13.9%, expressed as a percentage of the total volume, for the steady and unsteady state experiments, respectively. This can probably be explained by the water flux between the channel domain and the matrix, which is due to the hydraulic nonequilibrium. This increases the accessibility to the fast immobile domain and facilitates diffusion of the solute deeper into the matrix. The fraction of the fast immobile domain that is active in transferring solute can, accordingly, be expected to be larger in the unsteady state experiment.

4. Calibrating the Model

4.1. Steady State Experiment

The sample was at field capacity when the experiment began, so that $w_{im}^{fast} + w_{im}^{slow} = FC$. Since the matrix was at field capacity and the flow was steady, the system was in a state of hydraulic equilibrium. The S term is accordingly set to zero.

The diffusive mass transfer between the mobile and fast immobile domains is, as discussed in section 2.1, assumed to be sufficiently fast so that the domains can be regarded as perfectly mixed. The size of the fast immobile domain is not known, but by using the peak value of the BTC $\hat{C}$, the size of the fast immobile domain that is active in transferring solute could be determined from a mass balance as

$$fw_{im}^{fast} = [m/(V\hat{C})] - w_m \quad (27)$$

The transfer coefficient α was then adjusted and gradually increased to a point at which perfectly mixed conditions, according to the assumption above, just set in. This is the point at which the model fits the observed peak of the BTC, so $C_{im}^{fast}(t_{sf}) = C_m(z, t_{sf})$, $0 \le z \le Z$, where t_{sf} is the arrival time of the solute front. The result of the calibration procedure is shown in Figure 5 and is summarized below:

$$\begin{cases} \alpha = 1.5 \times 10^{-4}\ s^{-1} \\ \beta = 6.0 \times 10^{-6}\ s^{-1} \\ fw_{im}^{fast} = 0.057 \end{cases}$$

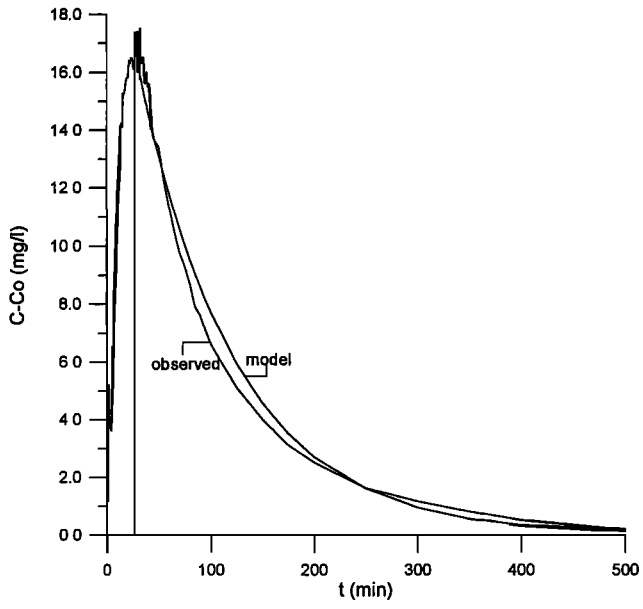

Figure 5. Measured BTC and calibrated model BTC.

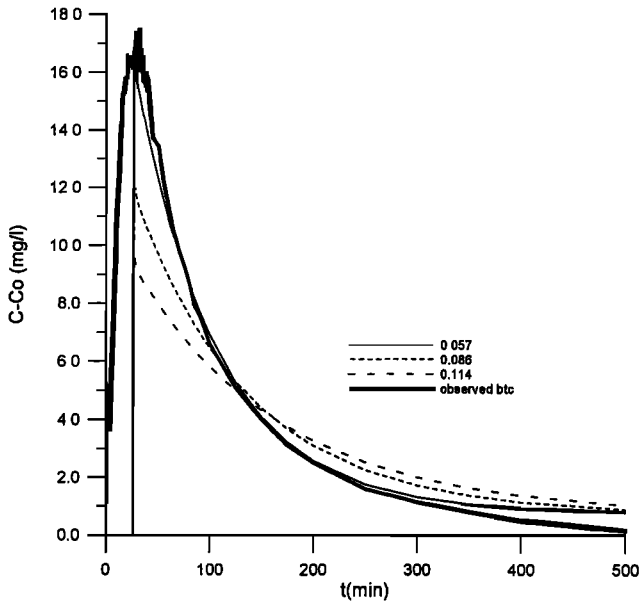

Figure 6. Demonstration of the effect of the size of fw_{im}^{fast} on the predicted BTC.

As can be seen in Figure 5, the model cannot describe the dispersion of the solute front because of the strictly convective solute flux law that was employed. The value of the diffusion coefficient β governing the diffusive transport between the fast and the slow immobile domain was adjusted to get the best possible fit between the model and the experimental BTC. Omitting the time period $0 < t < t_{sf}$, since the model cannot handle dispersion, the coefficient of determination was calculated to be 0.77 (61 data points).

The influence on the model result of the parameters α, β, and fw_{im}^{fast} was investigated by performing a sensitivity analysis. The effect of the size of the active part of the fast immobile domain on the modeled BTC is demonstrated in Figure 6. The size of the domain was increased by a factor of 1.5 ($fw_{im}^{fast} = 0.086$) and by a factor of 2 ($fw_{im}^{fast} = 0.114$). Keeping the other parameters constant and increasing the size of the active part of fast immobile domain will result in a decrease of the predicted peak of the BTC and, in the same way a decrease of the size of the domain will produce the opposite effect. In addition, because of the low concentration in the fast immobile domain, the BTC will drop momentarily when the solute pulse has ended. The diffusional mass transport out from the fast immobile domain gives a long tail on the BTC.

The parameter α has a significant importance for the predicted concentration peak of the BTC. Keeping the other parameters constant and decreasing the α parameter increases the predicted peak of the BTC in a nonlinear way, which is in accordance with (23). Predicted BTCs for three cases, $\alpha = 1.5 \times 10^{-4}\ s^{-1}$, $\alpha = 1.0 \times 10^{-4}\ s^{-1}$, and $\alpha = 5.0 \times 10^{-5}\ s^{-1}$ are shown in Figure 7. The parameter β governs the accumulation dynamics of solute in the slow immobile domain and influences mainly the recession part of the BTC. By increasing the β parameter, the resulting BTC declination becomes faster in the beginning and slower in the end as solute is transported back into the channels. In the same way, by decreasing β the predicted BTC declination becomes slower in the beginning and faster in the end. This is demonstrated in Figure 8.

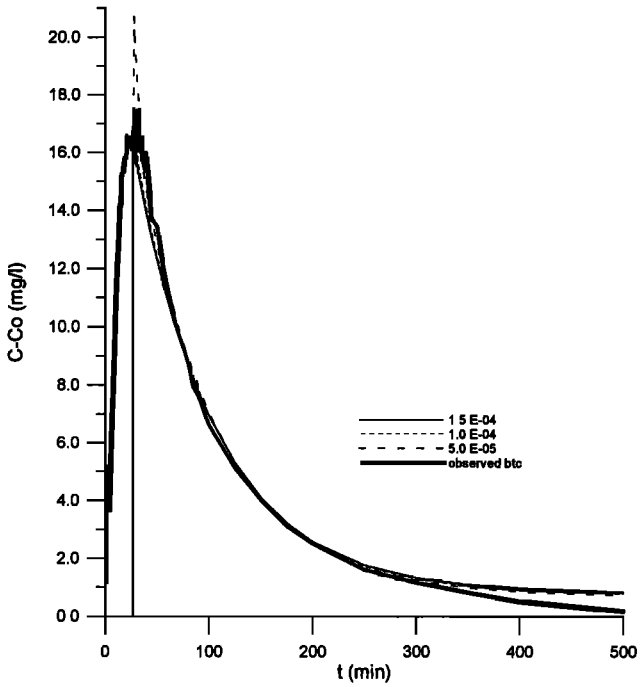

Figure 7. Sensitivity analyses with model parameter α.

4.2. Unsteady State Experiment

The system is here in a state of both physical and hydraulic nonequilibrium. The S term plays the role of an exchange term and switches signs depending on the direction of the hydraulic gradient between the matrix and the channel domain. The solution of the characteristic equations (6) and (7) has been presented by *Bendz et al.* [1998] for the case when a sequence of square pulses of volume flux density q_u with a duration T is supplied to the surface of the medium. The solution is divided into two main domains, the infiltration domain and the drainage domain, denoted as $D1$ and $D2$, respectively. A portion of the drainage domain forms a third solution domain, $D3$. The solution domains are shown in Figure 9.

In the infiltration domain $D1$, S is positive. Water is forced into voids and pores in the drainable region of the matrix because of the potential gradient that builds up during the irrigation pulses. The characteristics originate from the time axis on point t_s, where $0 \leq t_s \leq T$. In domain $D2$, water is flowing back into the channel system, and the S term becomes negative. Domain $D3$ is a region of internal drainage; however, the water potential is higher than in the storage region, so the S term is still positive. Domains $D1$ and $D3$ are divided by the drainage front that originates from the position T on the time axis. A moving boundary, originating from position T on the t axis, divides domains $D2$ and $D3$ and carries the water content at which the mobile and immobile regions are in a state of hydraulic equilibrium; that is, S is zero. The characteristics in $D2$ and $D3$ originate from the position T with a water content that varies from one characteristic to the other, from w_u to zero. The characteristics are therefore forming a fan shape.

Since the solute, according to (21), travels with the average water velocity, the characteristics of the solute transport differ from the characteristics of the water flow that were derived in our previous paper [*Bendz et al.*, 1998]. By inserting the expressions for the space and time history of the water content, given by the flow model, into (1) and employing a simple routing scheme using a Runga-Kutta algorithm, the solute characteristics were determined. The wetting front, the drainage front, and the moving boundary, $S = 0$, for the first and second pulses are shown in Figure 10 together with the solute characteristics.

According to the previous discussion it can be expected that the fraction of the fast immobile domain that was active in transferring solute was significantly larger under unsteady conditions than under steady state. This was verified by the model. By simply adjusting the size of the active part of the fast immobile domain to 0.094, a good fit to the experimental data was obtained. The coefficient of determination was found to be 0.94. In Figure 11 the observed and modeled BTCs are plotted for eight water pulses. Owing to the highly transient flow conditions here it is likely that the major part of the fast immobile

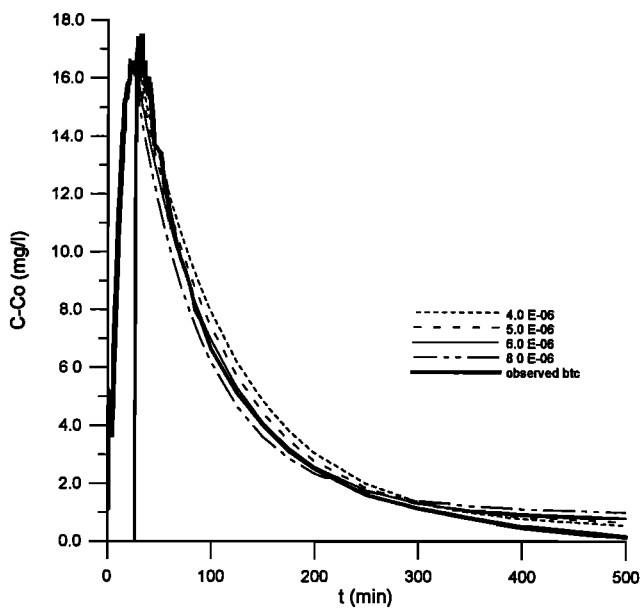

Figure 8. Sensitivity analysis with the model parameter β.

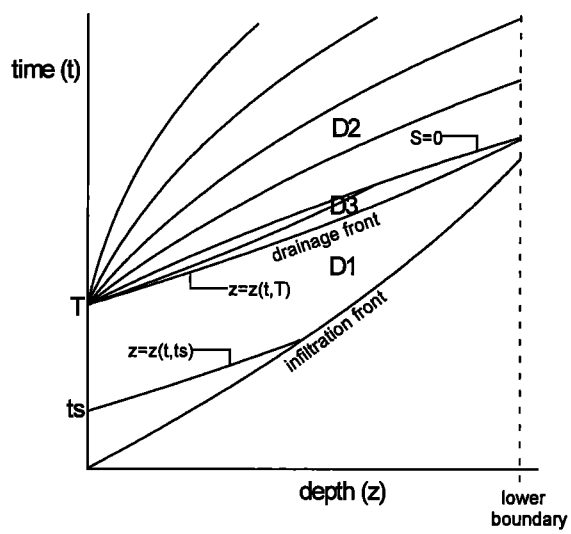

Figure 9. Solution domains for one water input event.

domain is active in transferring solute; in other words, f has approached 1.

5. Discussion and Conclusions

Several assumptions are built into the model, some of which have been made on an ad hoc basis and accordingly are only valid in this specific case. For example, it may be justified on a small scale to neglect the spatial variability by lumping the ensemble of channels into one channel domain and assuming that the observed dispersion of a solute pulse applied at the surface is solely the result of diffusional exchange of solute between the mobile domain and the immobile domains. However, on landfill scale the spatial variability must be taken into account. Other assumptions are made generally but are difficult to verify, and the experimental data can, at most, only indicate the soundness of the assumptions. Assumptions regarding the highly compacted and stratified nature of the waste medium and presence of impermeable surfaces (plastics), which promotes lateral flow at the expense of vertical matrix flow, have not been verified by measurements but are based on the first authors field observations during excavations of landfills. However, several field investigations have reported the existence of hanging water tables due to impermeable layers inside landfills, which supports the assumption [see, e.g., *Burrows et al.*, 1997; *Knox*, 1996].

Also, a good fit of the model to the observed data does not imply that the assumptions are correct. Conclusions regarding transport mechanisms cannot be drawn solely on the basis of the fitting of models to experimental data [*Brusseau and Rao*, 1990; *Jury and Roth*, 1990; *Sardin et al.*, 1991]. It is likely that the determined diffusional mass transfer coefficients, α and β, in the presented model also include effects of mechanical mixing and sorption processes.

Still, the proposed solute transport model together with the flow model presented by *Bendz et al.* [1998] is a first attempt to develop a conceptual understanding of the flow and transport processes in landfills. Prominent characteristics of the landfill waste medium have been identified, and their influence on the flow and transport processes has been taken into account. This is the essential quality of this work. The three-domain conceptualization appears to be physically sound, although it introduces parameters which are difficult to determine independently. The model has been shown to perform well as an interpretation tool despite the simple solute flux law. The proposed characteristic solution of the governing equation provides a didactic illustration of the solute flow paths. As long as a flow model is available which can describe the mobile water content in space and time, the proposed solution scheme is easy to apply for any boundary conditions with respect to water dynamics.

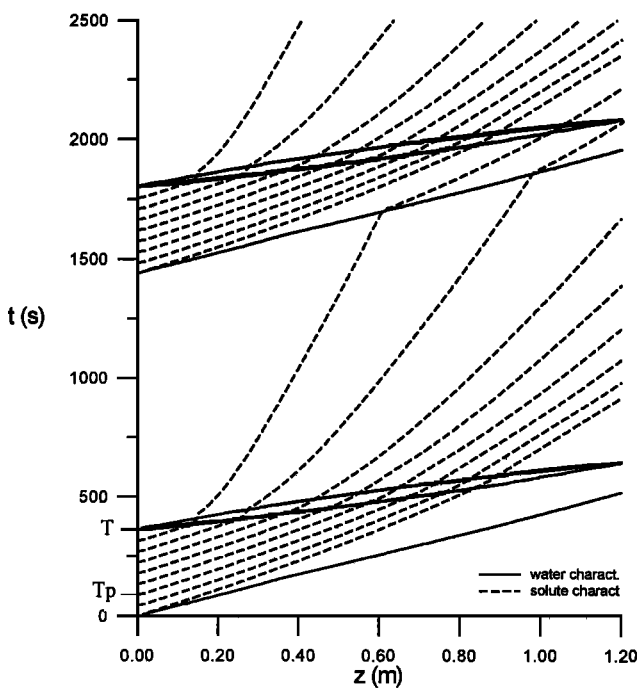

Figure 10. Solute characteristics plotted for an input of solute, duration T_p, followed by a sequence of square pulses.

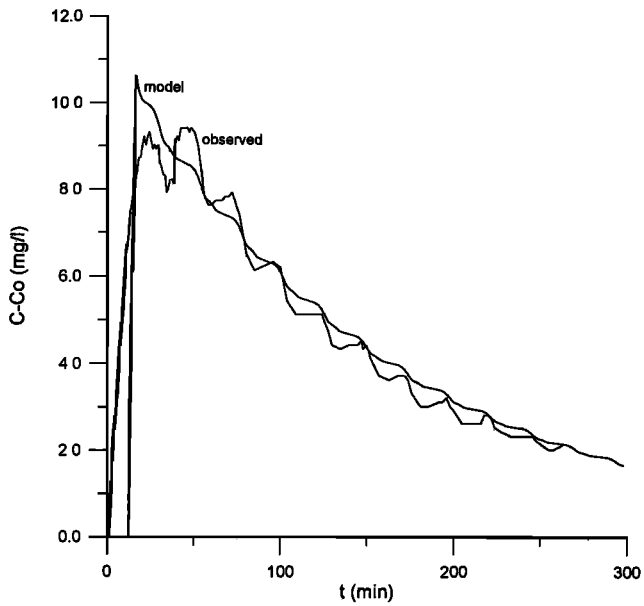

Figure 11. Observed and modeled BTCs.

It can be concluded that the residence time in the system is dominated by the accessibility to the fast immobile domain and the mass exchange between the immobile domains and the mobile domain. By applying the model to data from solute experiments performed under both steady and transient conditions, it has been demonstrated that the accessibility to the fast immobile domain is not fixed but is dependent on the water input pattern. The hydraulic gradients which develop between the mobile and the stagnant parts of the system increase the accessibility to the fast immobile domain so that a larger part of this domain becomes exposed to mobile water and thus becomes active in transferring solute. During highly transient conditions the major part of the fast immobile domain becomes active in transferring solute. In this investigation the size of the fast immobile domain can roughly be estimated to be about one tenth of the total waste volume. This result has significant practical implications for efforts to enhance the biodegradation process in landfills by recycling of the leachate and for strategies to flush out soluble contaminants. For these efforts to become successful, it is crucial to reach the whole pore volume of the landfill. This has been shown to be difficult because of the heterogeneity of landfill waste and the phenomenon of channel flow which shortcuts a large bulk of the land-

fill. In order to secure that as large portion as possible of the waste becomes exposed to water, it is favorable to create transient conditions in the landfill interior by irrigating the leachate in pulses.

The transport process in landfill waste has been shown in this investigation to be highly nonideal and in a state of physical and hydraulic nonequilibrium. This may imply that the observed leachate quality from landfills mainly reflects biochemical conditions in the channels and their surroundings. A weak leachate does therefore not imply that the landfill is stabilized and safely can be integrated in the environment without monitoring. A sudden settlement of the landfill, due to the biodegradation process, could change the geometry of the channel network. New parts of the landfill can be exposed to moving water, and, as a consequence, the leachate that is discharged at the lower boundary of the landfill may dramatically change its characteristics.

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D. Bendz, Department of Water Resources Engineering, Lund University, Box 118, 22100 Lund, Sweden. (David.Bendz@tvrl.lth.se)

V. P. Singh, Department of Civil and Environmental Engineering, Louisiana State University, Baton Rouge, LA 70803-7503. (cesing@unix1.sncc.lsu.edu)

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