

Analytical Mass Leaching Model for Contaminated Soil and Soil Stabilized Waste

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Abstract

An analytical model for evaluating mass leaching from contaminated soil or soil stabilized waste is presented. The model is based on mass transport due to advection, dispersion, and retardation and can be used to evaluate the suitability and/or efficiency of soil washing solutions based on the results of column leaching studies. The model differs from more traditional models for column leaching studies in that the analysis is based on the cumulative mass of leachate instead of leachate concentration. A cumulative mass basis for leaching eliminates the requirement for determination of "instantaneous" effluent concentrations in the more traditional column leaching approach thereby allowing for the collection of relatively large effluent volumes. The cumulative masses of three heavy metals—Cd, Pb, and Zn—leached from two specimens of soil mixed with fly ash are analyzed with the mass leaching model to illustrate application and limitation of the model.

Introduction

Leaching models have been used to evaluate admixture stabilization alternatives from the results of leaching tests. The results of static and dynamic leaching tests typically are analyzed using models based on the assumption of diffusion-controlled leaching behavior from the intact waste into the surrounding liquid with either instantaneous or kinetically controlled chemical reactions (e.g., Godbee and Joy, 1974; Johnson et al., 1979; Godbee et al., 1980; Nathwani and Phillips, 1978, 1980; Brown et al., 1986; Cote and Isabel, 1984; Cote and Bridle, 1987; Cote et al., 1987a, b; and Bishop, 1988).

Column leaching studies generally are evaluated qualitatively or in terms of empirical leaching models based on the total or relative leached quantity (mass) of contaminants. For example, several recent studies have evaluated the use of in situ soil washing technology for removing contaminants from polluted sites (Esposito et al., 1989; Hessling et al., 1990; Tuin and Tels, 1990a, b, 1991; Fountain et al., 1991; Hanson et al., 1993; O'Neill et al., 1993; Augustijn et al., 1994; van Benschoten et al., 1994; Davis and Singh, 1995; and Ozsu-Acar et al., 1995). The conclusions from these studies typically are based on the qualitative assessment of the total amount of contaminant mass leached to

evaluate the suitability and/or efficiency of a number of different extracting solutions (e.g., water, strong acids, chelating agents, surfactants) in removing a number of different contaminants (e.g., metals, radioisotopes, organic solvents) from soils with different properties.

In some cases, the results of column leaching tests have been evaluated in terms of traditional advective-dispersive transport theory. For example, Montera et al. (1994) describe the application of a numerical model to describe the leaching of residual copper (Cu) from tailings deposits with a strong acid solution (H_2SO_4). In their study, the dissolution of solid copper ore is modeled as a first-order decay process in which the decay constant is determined by fitting an analytical miscible transport model based on traditional advection-dispersion theory to the measured results of laboratory column leaching tests.

Although their study illustrates the potential use of analytical leaching models for practical applications, the analytical model utilized by Montera et al. (1994) is based on effluent concentrations of Cu. In general, no similar analytical model for evaluating the results of column leaching tests in terms of mass leached is available even though the majority of the results of soil washing studies are presented in terms of relative cumulative mass leached. Thus, the purpose of this paper is to present an analytical model for analyzing the results of column leaching tests for evaluating the suitability and/or efficiency of soil flushing or leaching from soil stabilized waste in terms of cumulative mass of the contaminant versus the contaminant concentration. The potential advantages of a model based on cumulative mass for evaluating the suitability and/or efficiency of a given extracting solution in soil flushing and leaching studies are emphasized.

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General Leaching Behavior

Limiting Case of Piston Flow

A general description of the proposed analytical mass leaching model relative to the more traditional concentration-based leaching model is provided to distinguish between evaluation of column leaching data based on solute mass or solute concentration. The limiting behavior of these two models is described by the case where the purely advective transport condition (i.e., no dispersion/diffusion), commonly referred to as piston or plug flow, governs the leaching of the waste material.

Concentration Basis for Piston Flow

In traditional column tests used to evaluate transport through porous media, complete breakthrough occurs when the effluent concentration, c_e , equals the influent concentration, c_o , for a given chemical species (i.e., $c_e/c_o = 1$) for the condition that c_o is maintained constant. For piston flow conditions, $c_e/c_o = 1$ occurs at $T = R_d$, where T is the pore volume of flow, and R_d is the retardation factor for linear and reversible instantaneous sorption (e.g., Freeze and Cherry, 1979). However, in the case of leaching a chemical species initially at a uniform equilibrium concentration in the pore phase of the medium, c_o , the elution curves (i.e., breakthrough curves for leaching) are represented by the complement of the solute breakthrough curves for traditional column tests (e.g., Brusseau, 1996). Thus, in the case of piston flow, the elution curves for leaching are described by the following equation:

$$\frac{c_e}{c_o} = \begin{cases} 1; & T' \leq R_d \\ 0; & T' > R_d \end{cases} \quad (1)$$

where T' is the leached pore volume of flow defined as follows:

$$T' = \frac{vt'}{L} \quad (2)$$

where v is the average linear fluid velocity, t' is the elapsed time, and L is the length of the column. Elution curves resulting from consideration of equation (1), which indicates a jump in c_e/c_o from 1 to 0 at $T' = R_d$, are shown in Figure 1a as a function of the retardation factor, R_d .

Mass Basis for Piston Flow Leaching from Contaminated Soil

An evaluation of the initial solute mass, m_o , in the pore fluid of the porous medium may be appropriate in the case where there is contamination within the soil that can be measured after squeezing the pore fluid from a soil sample recovered from the field. For example, high pressure squeezing to determine pore fluid concentrations in soil is a common practice for both laboratory and field diffusion testing of nonvolatile contaminants (Shackelford, 1991). In this case, the initial distribution of the contaminant between the liquid phase (pore fluid) and the solid phase (adsorbed) is not known a priori.

Thus, in this case, the initial solute mass, m_o , equals the product $V_p c_o$ where V_p equals one pore (void) volume of the porous medium, and the increment of solute mass Δm , leached in any given increment of time for piston flow conditions may be normalized with respect to m_o is given as follows:

$$\frac{\Delta m}{m_o} = \left(\frac{\Delta V_e}{V_p} \right) \left(\frac{c_e}{c_o} \right) \quad (3)$$

where ΔV_e is the effluent volume leached during the pore volume increment, $\Delta T'$, or

$$\Delta V_e = V_p \Delta T' \quad (4)$$

Substitution of equation (4) into equation (3) results in the following expression for the incremental leaching mass ratio for piston flow conditions:

$$\frac{\Delta m}{m_o} = \Delta T' \left(\frac{c_e}{c_o} \right) \quad (5)$$

which describes the increment of mass leached in a given leached pore volume increment.

The cumulative leaching mass ratio normalized with respect to the initial mass of the chemical species in the pore phase of the medium, LMR_m , represents the sum of all incremental masses leached from $T' = 0$ to $T' = \Sigma(\Delta T')$, or

$$LMR_m = \Sigma \frac{(\Delta m)}{m_o} = \Sigma (\Delta T') \left(\frac{c_e}{c_o} \right) \quad (6)$$

where the summation is over the number of effluent samples recovered sequentially since the beginning of the test upon which the value of LMR_m is based. Since there is a discontinuity in the effluent concentration ratio function at $T' = R_d$ [equation (1) and Figure 1b], c_e/c_o in equation (6) remains within the summation convention. Thus, for $0 \leq T' \leq R_d$, $c_e/c_o = 1$ [equation (1)], and equation (6) becomes

$$LMR_m = \Sigma (\Delta T') \left(\frac{c_e}{c_o} \right) = (T') (1) = T' \quad (7)$$

where the maximum value of LMR_m is R_d . For $R_d < T' < \infty$, $c_e/c_o = 0$, and equation (6) becomes

$$LMR_m = \Sigma (\Delta T') \left(\frac{c_e}{c_o} \right) = (R_d) (1) + (T') (0) = R_d \quad (8)$$

Equations (7) and (8) may be written more concisely as follows:

$$LMR_m = \Sigma \frac{(\Delta m)}{m_o} = \begin{cases} T'; & T' \leq R_d \\ R_d; & T' > R_d \end{cases} \quad (9)$$

Therefore, LMR_m for piston flow leaching results in a linear relationship between LMR_m and T' with a 1:1 slope until $T' = R_d$ followed by a constant value of $LMR_m = R_d$ for $T' \geq R_d$, as illustrated in Figure 1b.

Mass Basis for Piston Flow Leaching from Soil Stabilized Waste

When a soil is mixed with a liquid waste for the purpose of minimizing the leaching of contaminants from the waste (i.e., stabilizing the waste), some of the contaminant initially in the liquid waste may adsorb to the soil during the mixing process. Thus, the distribution of the contaminant between the waste and the soil after mixing may not be known although the total amount of contaminant mass in the liquid waste before mixing can be determined from knowledge of the volume of liquid waste, V_o , and the initial concentration of the contaminant, c_o , in the liquid waste. Thus, in this case, the total initial mass of a given contaminant, $M_o (= V_o c_o)$, is known even though the initial distribution between the solid phase mass (m_s) and liquid phase mass (m_o) after mixing the soil with the liquid is not known.

The leaching mass ratio as defined by equation (9) is with respect to the mass of contaminant initially within the pore fluid of the porous medium. However, the leaching mass ratio also may be normalized with respect to the total mass (i.e., adsorbed plus pore fluid phases) in the porous medium, $M_o (= m_o + m_s)$, by recognizing that m_o is related to M_o at equilibrium by the retardation factor as follows:

$$m_o = \frac{M_o}{R_d} \quad (10)$$

Thus, the incremental mass leached for piston flow normalized with respect to M_o is given by substituting equation (10) into equation (5), or

$$\frac{\Delta m}{M_o} = \left(\frac{\Delta T'}{R_d} \right) \left(\frac{c_e}{c_o} \right) \quad (11)$$

A similar analysis on equation (11) as performed on equation (3) for the case of piston flow leaching results in the following equation:

$$\text{LMR}_M = \sum \frac{(\Delta m)}{M_o} = \begin{cases} (T'/R_d); & T' \leq R_d \\ 1; & T' > R_d \end{cases} \quad (12)$$

where LMR_M is the cumulative leaching mass ratio normalized with respect to M_o . Thus, the cumulative leaching mass ratio, LMR_M , for piston flow will result in a linear relationship between LMR_M and T' with a slope of R_d^{-1} until $T' = R_d$ followed by a constant value of $\text{LMR}_M = 1$ for $T' \geq R_d$, as

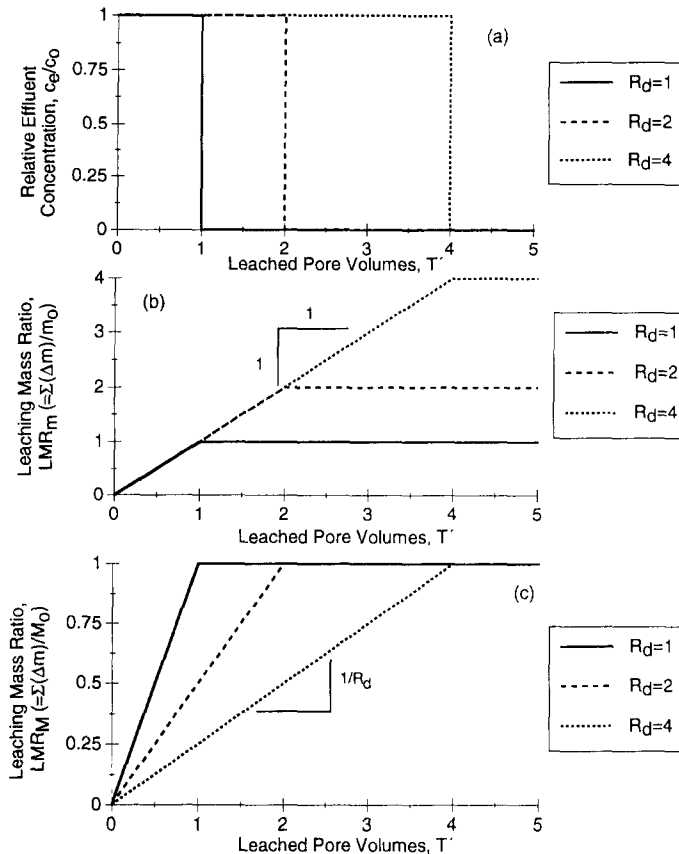


Fig. 1. Elution curves for piston flow behavior based on (a) relative effluent concentration, and cumulative leached mass normalized with respect to (b) initial contaminant mass in the soil pore water (m_o), and (c) initial total contaminant mass in the soil (M_o).

illustrated in Figure 1c. In general, equation (12) may be a more useful model than equation (9) for the case of a liquid waste admixed with soil.

General Case of Advective-Dispersive Transport

The general case of advective-dispersive transport during flushing or leaching is described by solutions to the advective-dispersive-reactive equation (ADRE) as follows (e.g., Brusseau, 1996):

$$R_d \frac{\partial c_r}{\partial t} = D \frac{\partial^2 c_r}{\partial x^2} - v \frac{\partial c_r}{\partial x} \quad (13)$$

where c_r is resident or pore fluid solute concentration within the porous medium, D is the dispersion coefficient including both mechanical dispersion and diffusion, x is distance, and t is time. Several analytical solutions to equation (13) have been derived depending on the initial and boundary conditions (e.g., van Genuchten, 1981; and van Genuchten and Alves, 1982).

Concentration Basis for Advective-Dispersive Leaching

A commonly applied analytical solution to equation (13) for the case of advective-dispersive leaching evaluated at $x = L$ is given as follows:

$$\frac{c_e(L, T')}{c_o} = 1 - \frac{1}{2} \left\{ \text{erfc} \left(\frac{R_d - T'}{2(T'R_d/P_L)^{1/2}} \right) + \exp(P_L) \text{erfc} \left(\frac{R_d + T'}{2(T'R_d/P_L)^{1/2}} \right) \right\} \quad (14)$$

where $P_L (= vL/D)$ is the column Peclet number, and erfc is the complementary error function. The complete derivation for equation (14) is given in the Appendix.

As in the case of piston flow, equation (14) represents the complement of the traditional solute breakthrough curves in the case of advective-dispersive transport through porous media. For example, the elution curves for advective-dispersive transport based on equation (14) are illustrated in Figure 2 for $P_L = 5, 10, 20,$ and 40 and for $R_d = 1$ and 4 . In general, the elution curves in Figure 2 correspond to the piston flow elution curves in Figure 1a except the spreading of the solute during transport due to dispersion is apparent. This dispersive tendency becomes more prevalent as the value for P_L decreases, i.e., as diffusion becomes more prevalent relative to advection. The effect of diffusion on the overall leaching rate also is evident by $T'_{0.5}$ (i.e., T' at $c_e/c_o = 0.5$) values $< R_d$ at low flow rates (e.g., see Shackelford, 1994). In the limit as $P_L \rightarrow \infty$, $T'_{0.5} \rightarrow R_d$; i.e., as advective transport increases relative to dispersive/diffusive transport (i.e., as P_L increases), the value of $T'_{0.5}$ approaches the value of R_d .

Mass Basis for Advective-Dispersive Leaching from Contaminated Soil

A mass basis for advective-dispersive leaching may be developed following the procedures outlined by Shackelford (1995a, b) for the cumulative mass approach to traditional column testing. In general, these procedures include (1) determining the local concentration gradient through spatial differentiation of the analytical concentration-based solution [i.e. $\partial c_r(x, t)/\partial x$], (2) substituting the result of step (1) along with the analytical solution [i.e., $c_r(x, t)$] into the equation for solute mass flux, $J(x, t) [= \theta v c_r(x, t) - \theta D \partial c_r(x, t)/\partial x]$, where θ is the

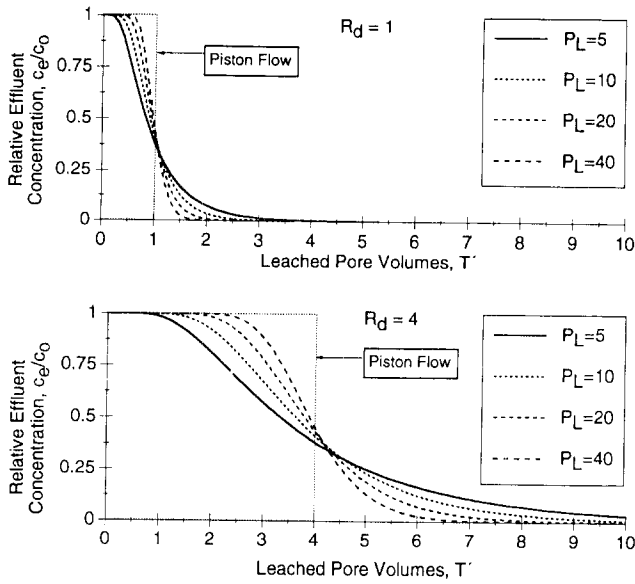


Fig. 2. Elution curves based on relative effluent concentration [equation (14)] for advective-dispersive transport illustrating the effect of the column Peclet number (P_L) for two different values of the retardation factor (R_d).

volumetric water content], and (3) integrating the resulting flux equation with respect to time to determine the cumulative amount of mass leached. The cumulative leached mass then is normalized with respect to the initial mass of contaminant in the porous medium. Application of the above procedure applied to the concentration-based analytical solution for resident concentrations [Appendix, equations (A3) and (A4)] for the case where the initial mass is defined with respect to the pore fluid in a contaminated soil, m_o , is given as follows:

$$\begin{aligned} \text{LMR}_m &= \sum \frac{(\Delta m)}{m_o} \\ &= T' - \frac{R_d}{2} \left\{ \left(\frac{T'}{R_d} - 1 \right) \text{erfc} \left(\frac{R_d - T'}{2(T'R_d/P_L)^{1/2}} \right) \right. \\ &\quad \left. + \left(\frac{T'}{R_d} + 1 \right) \exp(P_L) \text{erfc} \left(\frac{R_d + T'}{2(T'R_d/P_L)^{1/2}} \right) \right\} \quad (15) \end{aligned}$$

Elution curves based on equation (15) are shown in Figures 3 and 4. The effect on the cumulative mass leached of varying P_L for a given value of R_d is illustrated in Figure 3, whereas the effect of R_d for a given value of P_L on the cumulative mass leached is illustrated in Figure 4. The effect of increasing P_L is to result in a larger LMR_m value for a given value of T' at all values of R_d indicating a decrease in dispersive/diffusive transport relative to advective transport. A comparison of the cumulative mass leaching curves in Figures 1b with those in Figures 3 and 4 indicates that the LMR_m versus T' curves in Figures 3 and 4 approach the respective limiting elution curves for piston flow given in Figure 1b as $P_L \rightarrow \infty$.

Mass Basis for Advective-Dispersive Leaching from Soil Stabilized Waste

The cumulative mass for advective-dispersive leaching also may be normalized with respect to the total initial mass in the porous medium, M_o , simply by substituting equation (10) into

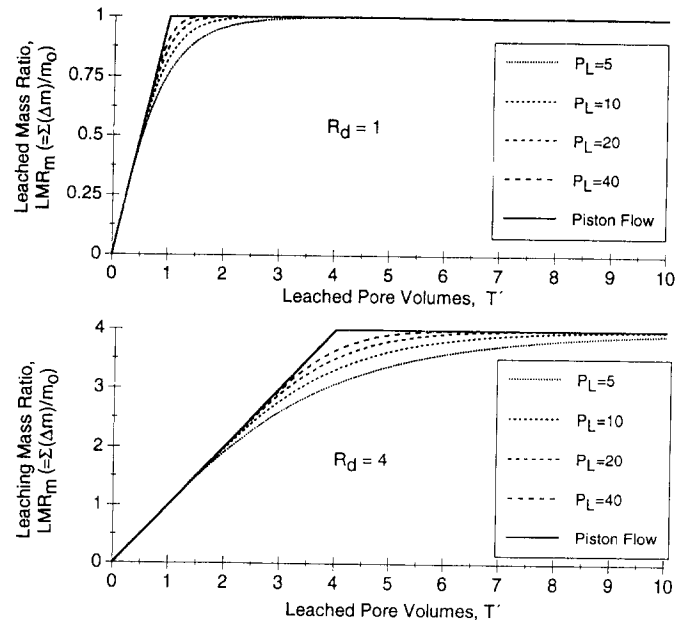


Fig. 3. Elution curves based on the leaching mass ratio approach normalized with respect to initial contaminant mass in pore water, m_o [equation (15)] illustrating the effect of the column Peclet number (P_L) for two different values of the retardation factor (R_d).

equation (15) resulting in the following definition of the leaching mass ratio, LMR_M :

$$\begin{aligned} \text{LMR}_M &= \sum \frac{(\Delta m)}{M_o} \\ &= \frac{T'}{R_d} - \frac{1}{2} \left\{ \left(\frac{T'}{R_d} - 1 \right) \text{erfc} \left(\frac{R_d - T'}{2(T'R_d/P_L)^{1/2}} \right) \right. \\ &\quad \left. + \left(\frac{T'}{R_d} + 1 \right) \exp(P_L) \text{erfc} \left(\frac{R_d + T'}{2(T'R_d/P_L)^{1/2}} \right) \right\} \quad (16) \end{aligned}$$

As with the situation for piston flow leaching, equation (16) for advective-dispersive leaching may be more useful than equation (15) for stabilization purposes where a soil is added to a liquid waste before mixing such that the initial distribution in contami-

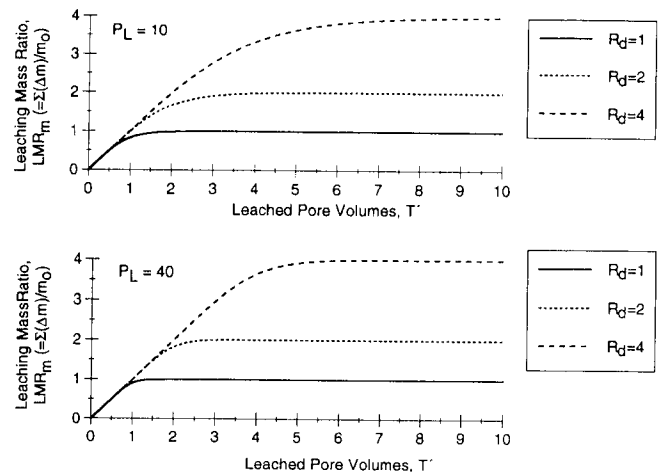


Fig. 4. Elution curves based on the leaching mass ratio approach normalized with respect to initial contaminant mass in pore water, m_o [equation (15)] illustrating the effect of the retardation factor (R_d) for two different values of the column Peclet number (P_L).

nant mass between solid and liquid phases after mixing is unknown.

The LMR_M curves based on equation (16) are illustrated in Figure 5 for varying P_L for a given value of R_d , and in Figure 6 for varying R_d for a given value of P_L . Again, the LMR_M versus T' curves in Figures 5 and 6 approach the respective limiting elution curves for piston flow [equation (12)] shown in Figure 1c as $P_L \rightarrow \infty$.

The Leaching Mass Ratio Approach

Based on the comparison of the elution curves using equation (14) versus equation (15) or equation (16), there are two approaches for quantitatively evaluating the leaching behavior

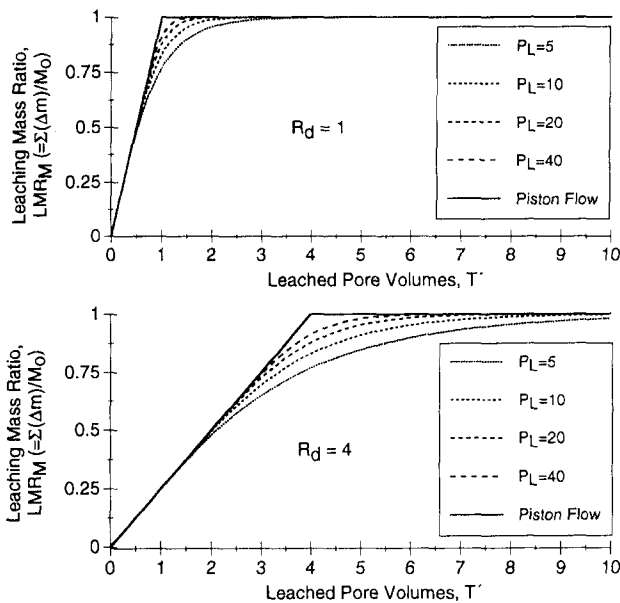


Fig. 5. Elution curves based on the leaching mass ratio approach normalized with respect to initial total contaminant mass in the soil, M_o [equation (16)] illustrating the effect of the column Peclet number (P_L) for two different values of the retardation factor (R_d).

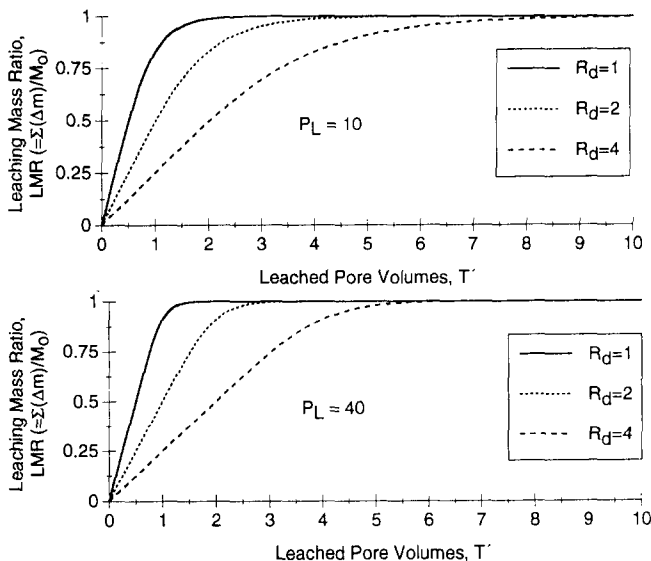


Fig. 6. Elution curves based on the leaching mass ratio approach normalized with respect to initial total contaminant mass in the soil, M_o [equation (16)] illustrating the effect of the retardation factor (R_d) for two different values of the column Peclet number (P_L).

of a contaminated soil or soil stabilized waste in terms of the proposed model based on traditional advective-dispersive transport theory. The first approach is to measure “instantaneous” effluent concentrations normalized with respect to the initial solute concentration in the pore water of the soil or stabilized waste, c_e/c_o , plot these values versus the leached pore volumes, T' , and apply equation (14) to the measured eluent curve to estimate P_L (or D) and R_d . The second approach is to (1) allow an increment of effluent, ΔV_e , to collect in an effluent reservoir over a given period, $\Delta T'$; (2) measure an average effluent concentration, \bar{c}_e , corresponding to the increment of the eluent mass, Δm , existing in this leached pore volume increment; (3) calculate $\Delta m (= \bar{c}_e \cdot \Delta V_e)$; and (4) normalize the increment in leached mass with respect to either the initial mass of the solute in the pore fluid of the test specimen, m_o , or the initial total mass of the solute in the test specimen, M_o . This procedure is repeated in successive increments, and the normalized increments in leached mass are summed to provide the cumulative leaching mass ratio values plotted versus the cumulative (elapsed) leached pore volumes, T' , corresponding to the actual elapsed time at which ΔV_e is collected. The resulting LMR_m or LMR_M versus T' data then are evaluated in terms of either equation (15) or equation (16), respectively, to provide the estimates of P_L (or D) and R_d .

A potential advantage of the leaching mass ratio approach relative to the concentration-based approach is related to the use of mass in the analysis versus the use of concentration. The continuum approach inherent in the derivation of equation (13) and, therefore, equation (14) implies that the c_e/c_o versus T' elution curves describe a continuous distribution of an infinite number of “instantaneous” concentrations. Since the measured effluent concentrations are a function of the effluent volume, only the collection of infinitesimally small volumes of effluent will allow for an accurate assessment of the effluent concentrations. Although the LMR_m or LMR_M versus T' elution curves also represent a continuous distribution of an infinite number of leaching mass ratio values, the influence of effluent sample volume has been removed from consideration since the LMR_m or LMR_M values are based on cumulative leached mass and, therefore, average effluent concentrations, \bar{c}_e , not instantaneous concentrations, c_e . Thus, the time-dependent distribution of c_e within the collection interval, $\Delta T'$, is not required in the leaching mass ratio approach allowing for collection of relatively large increments of effluent volume, $\Delta V_e (= \Delta T' \cdot V_p)$, and potentially less frequent sampling, beneficial aspects for long-term leaching studies.

Shackelford (1995b) illustrates the potential advantages associated with the measurement of cumulative effluent mass for traditional column testing by comparing the regressed values of P_L and R_d based on both effluent concentrations and effluent cumulative mass for column tests performed with a 0.01 M NaCl solution permeated through processed kaolin clay. The values of P_L and R_d regressed from the data plotted in terms of traditional relative effluent concentrations were essentially the same as the values of P_L and R_d regressed from the data plotted in terms of cumulative mass, with slight differences ($< 3\%$) in the regressed value of P_L being attributed to the error in plotting the average effluent concentrations at the middle of the sampling interval in the traditional approach. Shackelford (1995b) concludes that the cumulative mass procedure is more convenient and potentially more cost-effective than the concentration approach since the analysis can be based on the collection of relatively large volumes

of effluent thereby eliminating the need for more frequent effluent sampling. However, caution also should be exercised in the cumulative mass procedure since the loss (or gain) of mass in the collection reservoir over the extended periods of collection due to the existence of chemical (e.g., precipitation, volatilization) and/or biological (e.g., biodegradation) reactions may significantly affect evaluation of the leaching data.

The cumulative mass data also can provide a direct measurement of the retardation factor, R_d , i.e., without regression analysis (Shackelford, 1995b). In the case of the leaching mass ratio approach, $LMR_m [= \Sigma(\Delta m/m_o)]$ approaches R_d with increased leaching, as indicated in Figures 1b, 3, and 4, whereas the slope of the initial, tangent portion of the plot of $LMR_M [= \Sigma(\Delta m/M_o)]$ versus T' represents R_d^{-1} , as shown in Figures 1c, 5, and 6.

From a practical viewpoint, determining R_d from the extension of the steady-state portion of the $LMR_m [= \Sigma(\Delta m/m_o)]$ versus T' elution curve as per Figures 1b, 3, and 4 probably is more accurate since slight variations in the slopes of the $LMR_M [= \Sigma(\Delta m/M_o)]$ versus T' plots in Figures 1c, 4, and 5 will result in more significant differences in the estimated R_d values. However, the method of determining R_d illustrated in Figures 1b, 3, and 4 requires that steady-state leaching conditions be established, whereas the steady-state conditions are not necessarily required to provide an estimate of the initial (tangential) slope of the $LMR_M [= \Sigma(\Delta m/M_o)]$ versus T' plots in Figures 1c, 4, and 5.

Illustrative Application

An example of the use of the LMR_M model is illustrated with respect to the leaching of metals from two specimens of soil mixed with fly ash. Fly ash is a by-product of the coal combustion process. Fly ash has been evaluated for use as a binder admixture for stabilizing waste (Cote et al., 1987b; Roy et al., 1991). However, a potentially detrimental aspect of the use of fly ash in such applications is the leaching of toxic heavy metals. In this regard, fly ash also has been evaluated as a waste material (e.g., Young et al., 1984; Bowders et al., 1987, 1990; Creek and Shackelford, 1992; Edil et al., 1992; Usmen et al., 1992).

In the present situation, the application of the LMR_M model for evaluating the leaching of cadmium (Cd), lead (Pb), and zinc (Zn) from the results of column leaching tests performed on soil mixed with fly ash is illustrated. The soils used as the admixture materials are sand and bentonite. The fly ash was obtained from the Nixon Power Plant in Colorado Springs, CO, and is classified as a Class F fly ash. Based on the analysis by Creek (1990), the fly ash has a measured cation exchange capacity, CEC, of 6 meq/100 g, although the mechanism for this measured CEC is unknown. The sand is a clean, medium, 20-40 silica sand obtained from the Colorado Lien Company in LaPorte, CO. The bentonite is a sodium bentonite obtained from the Black Hills, WY, and has a measured CEC of 76 meq/100 g. Further details of the physical and chemical properties of the three constituent materials are provided by Shackelford and Glade (1994).

The results of two column leaching tests performed on two different test specimen mixtures are analyzed using the test procedure for the cumulative leaching mass ratio approach and the LMR_M model. Each test specimen contained 50 percent (dry weight) fly ash to provide a consistent basis for evaluating the potential leaching of the metals. However, the first test specimen

was admixed with 50 percent sand whereas the second test specimen was admixed with a 43.75 percent sand and 6.25 percent bentonite mixture to evaluate the potential effect of bentonite addition on reducing the leaching rate of the metals.

The test specimens were prepared by mixing the air-dried constituent materials with distilled water (DW) until a moisture content slightly above optimum moisture content based on the standard Proctor procedure (ASTM D 698) was achieved. The test specimen mixtures then were compacted into half-size compaction molds (volume = 472 cm³, length = 5.82 cm) according to the standard Proctor procedure (ASTM D 698) and cured for one week before being extruded and placed in flexible-wall permeameters. After assembling the flexible-wall permeameters, the test specimens were back-pressured to increase the initial degree of saturation before permeation. Further details of the test specimen preparation as well as the initial and final properties of the test specimens are given by Shackelford and Glade (1994).

Each test specimen was permeated with DW using a Harvard Apparatus flow pump (Model No. 944) described by Redmond and Shackelford (1994) and the permeation procedure described by Shackelford and Glade (1994). The flow pump apparatus is a mechanical device that is capable of maintaining constant, low flow rates for extended durations. In the present study, the flux of DW through the specimens was maintained constant at a rate of 1.37×10^{-7} m/s throughout the duration of the tests. The effluent from the column tests was allowed to accumulate in quarter pore volume increments ($\Delta T' = 0.25$) before removal for measurement of Cd, Pb, and Zn concentrations. A total of three pore volumes of effluent was collected in each test. Aliquots were recovered from each quarter pore volume increment (i.e., $\Delta V_e = 0.25V_p$), and the metal concentrations were measured using ICP-OES (Inductively Coupled Plasma-Optical Emissions Spectroscopy). The measured metal concentrations were used to calculate the leached mass of the metals, and the values were normalized with respect to the estimated total mass of each metal available in the constituent materials based on the extraction procedures described by Shackelford and Redmond (1995).

Nonlinear least-squares regression analyses were performed with equation (16) on the measured LMR_M values for Cd, Pb, and Zn versus T' from the two test specimens containing fly ash using the IGOR software program distributed by WaveMetrics, Inc. (Lake Oswego, OR). The parameter values for each regression analysis are shown in Table 1, and the measured data and regressed (theoretical) curves are shown in Figure 7. The analyses of the results from the test with the sand admixed fly ash (Test No. 1) indicates that each metal is reactive ($R_d > 1$). This reactivity can be attributed, in part, to the measured CEC of the fly ash constituting 50 percent of the specimen mixture. Although the mechanism for the measured CEC of the fly ash is unknown, the mechanism probably is not associated with a charge deficiency consistent with isomorphic substitution in clay mineral soils, such as the bentonite. Thus, the assumption of linear, reversible, and instantaneous sorption associated with the developed model may not be strictly applicable in the case of the fly ash. The fact that the model overestimates the initial eluted mass of all three metals for the Test No. 1 results, as shown in Figure 7, probably reflects, in part, a lack of strict conformance of the measured data with the model assumptions. Brusseau (1996) has noted similar differences for the case where a

Table 1. Regressed Transport Parameters from Column Leaching Tests

Test No. (1)	Mixture constituents (% by weight) ¹			Flow rate, <i>q</i> (m/s) (5)	Seepage velocity, <i>v</i> (m/s) (6)	Metal (7)	Regressed parameters (value ± std. dev.) ²		Dispersion coefficient, <i>D</i> (m ² /s) (10)	Extrapolated pore volumes required for 100% removal, <i>T'</i> ₁₀₀ (11)
	FA (2)	S (3)	B (4)				<i>P_L</i> (8)	<i>R_d</i> (9)		
1	50	50	—	1.37×10^{-7}	4.74×10^{-7}	Cd	2.70 ± 0.56	1.79 ± 0.12	1.02×10^{-8}	9.40
						Pb	0.658 ± 0.196	2.66 ± 0.36	4.19×10^{-8}	45.6
						Zn	0.984 ± 0.317	1.83 ± 0.27	2.80×10^{-8}	21.9
2	50	43.75	6.25	1.37×10^{-7}	4.74×10^{-7}	Cd	26.3 ± 6.6	5.50 ± 0.03	0.944×10^{-9}	8.90
						Pb	8.53 ± 1.79	5.33 ± 0.04	2.91×10^{-9}	13.5
						Zn	2.29 ± 0.28	3.94 ± 0.10	10.8×10^{-9}	23.3

¹FA = fly ash; S = sand; B = bentonite.

²*P_L* = column Peclet No. (= *vL*/*D*); *L* = column length (= 5.82 cm); *R_d* = retardation factor.

concentration-based analytical solution to equation (13) was used to predict the flushing of diethylether with water in an induced-gradient field test. Nonetheless, the increase in the retardation factor for each metal in Test No. 2 relative to Test No. 1 can be attributed, in part to the increase in the adsorptive capacity of the sand-bentonite admixture due to the bentonite addition relative to the pure sand admixture.

Also shown in Table 1 are the extrapolated pore volumes of leaching solution, *T'*₁₀₀ (to three significant figures), required to

achieve 100 percent removal efficiency of the metals based on the regressed values of *P_L* and *R_d*. Based on the *T'*₁₀₀ values for Cd and Pb in Table 1, the increase in retardation factor for Test No. 2 relative to Test No. 1 does not necessarily result in a greater time required to reach 100 percent removal efficiency. In the case of Cd and Pb, the significantly greater dispersion coefficients, *D*, associated with the results of Test No. 1 more than compensate for the increase in retardation factor due to the bentonite addition for Test No. 2. Thus, as indicated by this example, one utility

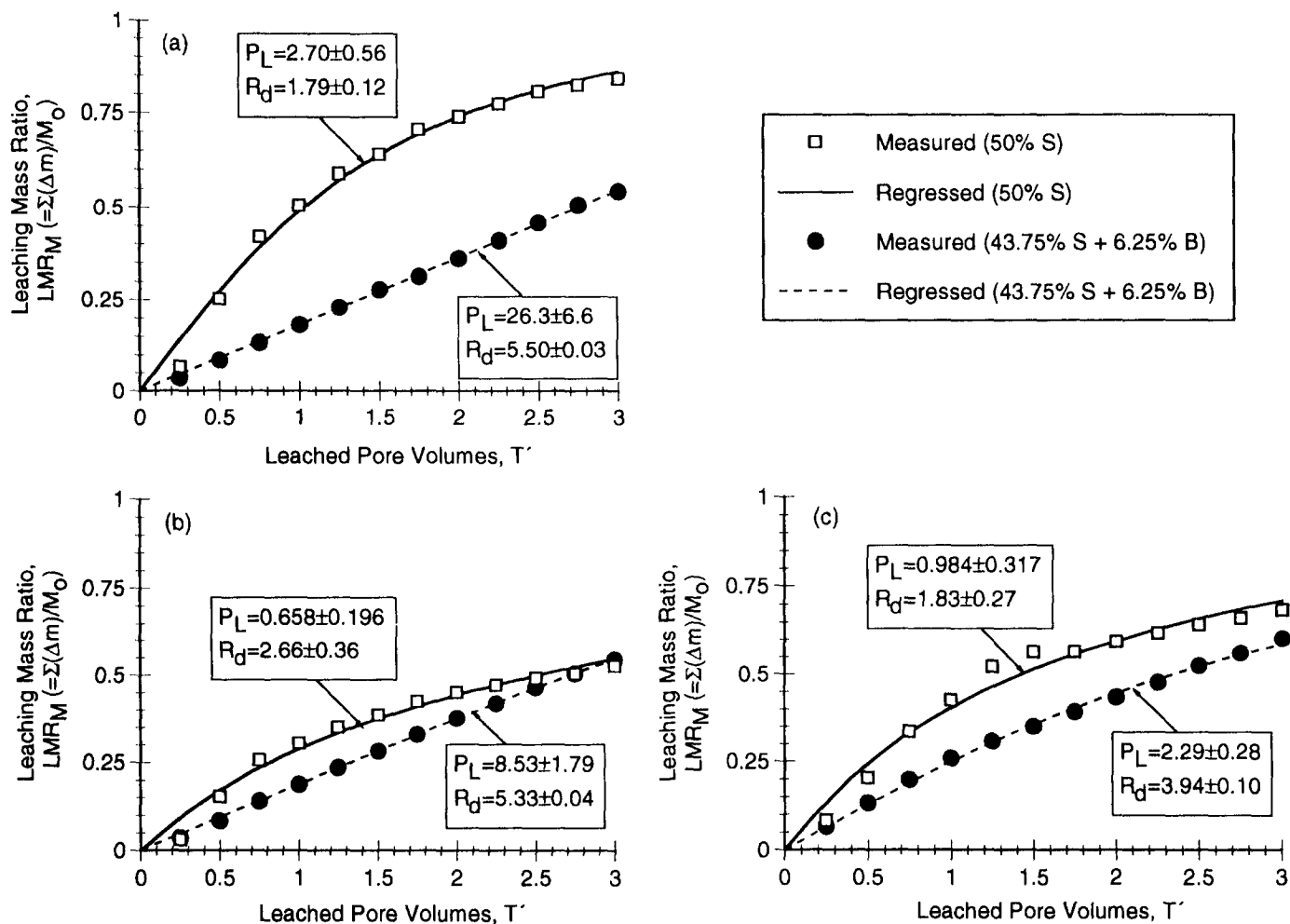


Fig. 7. Measured elution curves for 50 percent fly ash specimens mixed with 50 percent (dry wt.) soil admixtures (S = sand, B = bentonite) regressed with the leaching mass ratio model [equation (16)] for (a) cadmium (Cd), (b) lead (Pb), and (c) zinc (Zn).

of the leaching mass ratio approach is to provide a quantitative comparison for the extended performance of a column leaching study based on the short-term results from leaching tests.

Practical Applications and Limitations

In addition to evaluating the suitability and/or efficiency of a soil admixture in stabilizing a waste, the leaching mass ratio approach also may be used to evaluate the leaching of miscible chemical species from soils for any situation where there is solvent flow. Examples of potential applications include evaluating the potential effectiveness of pump and treat remediation activities for cleanup of contaminated sites and evaluating the efficiency of different chemical solutions in removing adsorbed contaminants from soils for applications involving in situ injection-withdrawal systems. However, the assumptions of instantaneous, linear, and reversible sorption may not be strictly applicable in the case where the chemical environment is not sufficiently time invariant. In such applications, the leaching mass ratio approach may be used to provide an indication of the overall mechanisms involved in the leaching process, but only a more detailed model which includes all possible transport processes and chemical reactions can provide a complete mechanistic description of the leaching process (e.g., see Rubin, 1983).

Summary and Conclusions

An analytical model for evaluating mass leaching from laboratory soil columns is presented. The model is based on mass transport due to advection, dispersion, and retardation and can be used to evaluate the suitability and/or efficiency of extracting solutions for soil flushing from the results of laboratory leaching tests. The model differs from more traditional models for column leaching studies in that the analysis is based on the leached cumulative mass instead of leached concentration. A cumulative mass basis for leaching allows the collection of relatively large effluent samples thereby eliminating the requirement for determination of "instantaneous" effluent concentrations in the more traditional column leaching approach.

The results of a laboratory column study used to evaluate the leaching of cadmium, lead, and zinc from fly ash stabilized with sand and sand-bentonite admixtures are evaluated with the mass leaching model to illustrate application and limitation of the test procedure and data analysis. The results of the analyses indicate that bentonite addition to an otherwise sand-fly ash mixture results in delayed leaching of all three metals. This delayed leaching behavior is attributed, in part, to the increase in adsorptive capacity of the soil admixture resulting from the bentonite addition.

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Appendix

The concentration-based analytical solution given by equation (14) represents a solution to equation (13) and the following initial and boundary conditions (van Genuchten, 1981; van Genuchten and Alves, 1982):

$$c_r(x, 0) = c_i$$

$$\left(v c_r - D \frac{\partial c_r}{\partial x} \right) \Big|_{x=0} = \begin{cases} v c_o; & 0 < t \leq t_o \\ 0; & t > t_o \end{cases} \quad (A1)$$

$$\frac{\partial c_r}{\partial x}(\infty, t) = 0$$

where c_o is the concentration of a given chemical species in the influent reservoir of a laboratory column which is assumed to be well-mixed; and c_i is the initial concentration of the same chemical species in the pore fluid of the porous medium. The upper or first boundary condition corresponds to a pulse, or plug, source concentration input in which a solution containing a solute at concentration, c_o , is permeated through the influent end of a column of porous medium continuously from time $t = 0$ to time $t = t_o$ followed immediately by a continuous flushing of the column with a solution with zero concentration of the solute for the remainder of the test (i.e., $t > t_o$). The general solution to equations (13) and (A1) may be written as follows (van Genuchten, 1981; van Genuchten and Alves, 1982):

$$c_r(x, t) = \begin{cases} c_i + (c_o - c_i) f(x, t); & 0 < t \leq t_o \\ c_i + (c_o - c_i) f(x, t) - c_o f(x, t - t_o); & t > t_o \end{cases} \quad \dots (A2)$$

where the function $f(x, t)$ varies from zero to unity [i.e., $0 \leq f(x, t) \leq 1$] and is given by (Lindstrom et al., 1967; Gershon and Nir, 1969; van Genuchten, 1981; and van Genuchten and Alves, 1982);

$$f(x, t) = \frac{1}{2} \left\{ \operatorname{erfc} \left(\frac{R_d x - vt}{2(DR_d t)^{1/2}} \right) + 2 \left(\frac{v^2 t}{\pi DR_d} \right)^{1/2} \exp \left[- \left(\frac{R_d x - vt}{2(DR_d t)^{1/2}} \right)^2 \right] - \left(1 + \frac{vx}{D} + \frac{v^2 t}{DR_d} \right) \exp \left(\frac{vx}{D} \right) \operatorname{erfc} \left(\frac{R_d x + vt}{2(DR_d t)^{1/2}} \right) \right\} \quad (A3)$$

For $t > t_o$, a modified time $t' (= t - t_o)$ is substituted for t in equation (A3) to evaluate the function $f(x, t') = f(x, t - t_o)$. Thus, $f(x, t')$ also varies from zero to unity [i.e., $0 \leq f(x, t') \leq 1$].

When t_o is sufficiently long such that $f(x, t) = 1.0$, equation (A2) for $t > t_o$ reduces to

$$c_r(x, t) = c_o - c_o f(x, t') \quad (A4)$$

This general solution, equation (A4), represents a decrease in solute concentration from c_o to zero for $t' \geq 0$. Thus, a value of $f(x, t) = 1.0$ is equivalent to the condition where the concentration, $c_r(x, t)$, is uniformly distributed throughout the pore fluid of the porous medium at an equilibrium value of c_o at time t_o . Therefore, equation (A4) can be considered as the general form of a leaching model in which there is a decrease in concentration from an initial concentration, c_o , to zero at a specified location for $t' \geq 0$.

Equations (A1)-(A4) are written in terms of resident, or pore-water concentrations. Based on studies by van Genuchten and Parker (1984) and others, the concentration of interest with respect to the measurement of solute breakthrough curves from finite columns of porous media is the flux-averaged effluent concentration, c_e , defined as the mass of solute passing through the effluent end of a finite soil column per unit volume of fluid during an elementary time interval, or:

$$c_c = \frac{J(L^-, t)}{q} = \frac{\theta}{q} \left[v c_r(L^-, t) - D \frac{\partial c_r(L^-, t)}{\partial x} \right] \quad (\text{A5})$$

where θ is the volumetric water content in the porous medium; q is the fluid flux given by Darcy's law; and $J(L^-, t)$ is the solute mass flux evaluated just inside the effluent end of the soil column. Evaluation of the effluent concentration in accordance with equation (A5) at time, t' , using the general solution given by equations (A3) and (A4), provides the following expression for the relative effluent concentration, c_c/c_o (van Genuchten and Parker, 1984; Shackelford, 1994):

$$\frac{c_c(L, t')}{c_o} = 1 - \frac{1}{2} \left\{ \operatorname{erfc} \left(\frac{R_d L - vt'}{2(DR_d t')^{1/2}} \right) + \exp \left(\frac{vL}{D} \right) \operatorname{erfc} \left(\frac{R_d L + vt'}{2(DR_d t')^{1/2}} \right) \right\} \quad (\text{A6})$$

where the negative sign on L has been ignored for convenience. Equation (A6) represents a leaching model in terms of effluent concentrations for a finite column of porous material in which there is a decrease in relative effluent concentration, c_c/c_o , from unity to zero for $t' \geq 0$.

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