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Unsaturated Moisture and Radionuclide Transport: Laboratory Analysis and Modeling

G. W. Gee
A. C. Campbell
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June 1981

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UNSATURATED MOISTURE AND RADIONUCLIDE
TRANSPORT: LABORATORY ANALYSIS
AND MODELING

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SUMMARY

This report describes several laboratory procedures and computer model simulations used to evaluate the transport of water and radionuclides through unsaturated Hanford soils.

The unsaturated hydraulic conductivity was measured using the steady state methods of Klute (1972) and the transient state method of Rose (1968). These experimental data were compared to the conductivity models of Millington and Quirk (1961); Davidson et al. (1969); Campbell (1974); van Genuchten (1980a); and Bresler, Russo and Miller (1978). Good agreement was found between all methods in the wet range; however, disagreement was found in the dry range. The method of Davidson et al. (1969) overpredicted the conductivity by several orders of magnitude in the <10% volumetric moisture content range. The other methods underpredicted the conductivities in this range. The power function approach of Campbell (1974), and Bresler, Russo and Miller (1978) best predicted the measured conductivities. However, none of the conductivity models explicitly addresses the water vapor component of the conductivity. This may explain the underprediction of the hydraulic conductivity in the dry range where vapor transport is important.

Radionuclide transport through unsaturated media was investigated by using two solute transport models to describe the transport of tritium and strontium-85 in laboratory columns. A two parameter convective-dispersive model was compared with a four parameter mobile-immobile water model (van Genuchten and Wierenga 1976a). Both models adequately described the movement of tritium and strontium through small (5 cm x 27.5 cm) columns and the movement of tritium through a large (0.5 m x 1.7 m) column.

The dispersion coefficient was found to be sensitive to changes in both velocity and column length. The mobile-immobile water equations were not as sensitive to changes in experimental scales as the convective-dispersive equation.

Both models were relatively successful in describing the rapid flush of strontium-85 from a column initially leached with a low salt solution followed

by a high salt solution, a phenomena called the snow plow effect. The four parameter mobile-immobile water model predicted the initial release of the strontium more accurately than the two parameter convective-dispersive model. Both models confirm enhanced mobility of strontium-85 with leaching solutions of increased salt concentration.

CONTENTS.

SUMMARY	iii
1.0 INTRODUCTION	1.1
2.0 BACKGROUND	2.1
2.1 UNSATURATED CONDUCTIVITY MODELS	2.1
2.2 RADIONUCLIDE TRANSPORT	2.5
3.0 MATERIALS AND METHODS	3.1
3.1 SOILS	3.1
3.2 SOLUTIONS	3.1
3.3 CONDUCTIVITY MEASUREMENTS	3.1
3.4 RADIONUCLIDE TRANSPORT TESTS	3.2
3.4.1 Large Column Tests	3.2
3.4.2 Snow Plow Effects	3.2
3.4.3 Equations Used for Computer Simulations	3.5
4.0 RESULTS AND DISCUSSION	4.1
4.1 UNSATURATED CONDUCTIVITY MEASUREMENTS	4.1
4.2 RADIONUCLIDE TRANSPORT MODELING	4.6
4.2.1 Curve Fitting Procedure	4.6
4.2.2 Small Column Experiments	4.6
4.2.3 Large Column Experiments	4.7
4.2.4 Snow Plow Effect	4.12
5.0 CONCLUSIONS	5.1
REFERENCES	R.1

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FIGURES

1	Model of Large Laboratory Layered Soil Column	3.3
2	Apparatus for Measuring Radionuclide Movement in Unsaturated Soil Columns	3.4
3	Calculated and Measured Hydraulic Conductivities, Campbell and BRM Method	4.2
4	Calculated and Measured Hydraulic Conductivities, BRM, MQ and Davidson et al. Methods	4.3
5	Hydraulic Conductivity Determined by Method of van Genuchten from Wetting and Drying Water Characteristics of Rupert Sand Compared With Steady State Experimental Data	4.4
6	⁸⁵ Sr Breakthrough Curve Using Small Column and High Salt Solution	4.8
7	Tritium Breakthrough Curve Using Small Column and High Salt Solution	4.9
8	Comparison of Tritium Breakthrough Curve Using Small and Large Column	4.10
9	Comparison of Two and Four Parameter Models for Large Column Tritium Breakthrough Curve	4.11
10	Comparison of Two and Four Parameter Models for Snow Plow Effect.	4.13
11	Sensitivity of Four Parameter Model to the Parameter f	4.14

TABLES

1	Parameter Values Estimated by Curve Fitting Routine for Small and Large Column Experiments	4.7
2	Water Content Versus Depth for the Large Column During Tritium Experiments	4.12

1.0 INTRODUCTION

Disposal practices for low level radioactive waste generally rely on the ability of soil and sediments to sorb radionuclides and isolate them from the rest of the biosphere. Considerable work has been done in the past to characterize sediments for their radionuclide sorption with little attention paid to the hydraulic properties of these materials. There is a growing understanding and appreciation of the role that moisture flow through sediments plays in the transport of radionuclides. In recent years more research has been conducted to characterize the moisture flow properties of Hanford sediments, particularly for unsaturated flow, since this is the dominant flow process occurring in arid shallow land burial sites (Jones 1978, Jones et al. 1979, Gee and Simmons 1979, Sisson and Gibbs 1979). This report describes some of the current efforts to combine knowledge of moisture flow and radionuclide sorption to predict contaminant transport. Problems discussed here include the measurement and prediction of the unsaturated hydraulic conductivity, and the computer modeling of one dimensional, unsaturated transport of radionuclides through sediments.

2.0 BACKGROUND

Accurate assessments of radionuclide transport in arid region burial grounds require application of the physics of unsaturated water flow to the transport of the radionuclides under investigation. Radionuclides in contact with burial ground soils will be transported only when the proper mechanisms are operational. The maximum rate of movement is limited by the molecular diffusion rates of the radionuclides and the convective action of the soil solution. The sorption characteristics of the soil material, subject to the solution-soil pH, Eh, and ionic composition, act to retard radionuclide movement, hence the radionuclides will generally move at a rate less than the soil solution. In arid regions where water tables are normally well below the bottom of the burial trench, unsaturated conditions prevail and the soil solution is transported under forces associated with surface evaporation and deep drainage. The transport of waste to the soil surface or to the water table over a long period of time is determined by the direction and rate of flow of the soil solution and the radionuclide adsorption/desorption characteristics of the soil. A conservative description of radionuclide transport in unsaturated soil is to assume that the radionuclide undergoes no sorption and movement can be estimated by characterizing the water flow using the best available technology. The parameters needed for this kind of assessment include the unsaturated water flow (hydraulic) properties of the soil, precipitation records and estimates of actual evaporation. A modeling effort for unsaturated flow using these input parameters (Gee and Simmons 1979) suggests that poorly sorbed radionuclides could either be transported to the soil surface or carried to the water table depending upon various climatic conditions. Prediction of moisture flow rates from climatic conditions requires a reasonable description of the hydraulic conductivity which is functionally dependent upon the water content and the energy state or capillary pressure of the soil water.

2.1 UNSATURATED CONDUCTIVITY MODELS

Knowledge of the hydraulic properties of the soil are key to the understanding of the transport of the water (soil solution) and hence of the radionuclide. Because of experimental difficulties, unsaturated conductivities are

often estimated from one or more models that are available in the literature. Various models have been used for predicting the hydraulic conductivity of unsaturated soils. Two main groups have been indentified by Mualem (1976). The first group assumes that the relative conductivity, K_r , is a power function of the effective saturation, S_e , i.e.,

$$K_r = \frac{K}{K_s} = S_e^a \quad (1)$$

where: K and K_s are the unsaturated and saturated conductivity respectively, $S_e = (\theta - \theta_r)/(\theta_s - \theta_r)$, where θ , θ_r and θ_s are the actual, residual, and saturated water contents, respectively, and a is an empirical parameter determined for each soil. The residual water content is not a well defined parameter but is qualitatively defined to be the water content at which the conductivity drops to an insignificantly low value; i.e., $K_r \approx 0$.

The second group of models makes use of the measured pressure head-water content curve (i.e., desorption curve, soil characteristic curve, etc.), to derive the unsaturated hydraulic conductivity. The important developments in this second group of models have come from Burdine (1953), Childs and Collis George (1950) and the modifications of these models by Millington and Quirk (1961). A general expression for this type model is:

$$K_r = S_e^b \frac{\sum_{i=1}^{\ell} \left(\frac{2(\ell-i) + 1}{h_i^2} \right)}{\sum_{i=1}^m \left(\frac{2(m-i) + 1}{h_i^2} \right)} \quad (2)$$

where m is the total number of pore classes, ℓ is the number of pore classes up to a prescribed value of θ , and h is the pressure head. The coefficient b represents a pore interaction factor that varies from 0 to 4/3 depending on the formulation (Green and Corey 1971).

Mualem (1976) has shown that similarity exists between equation (1) and (2) when the summation expression (2) is expressed in integral form. A somewhat similar approach has been used by Campbell (1974) to arrive at an expression identical in form to equation (1). His expression is

$$K_r = S_e^{2b + 3} \quad (3)$$

where b is the slope of water content pressure head curve plotted on log-log paper for a range of negative pressure heads below the air entry pressure.

van Genuchten (1980a) has developed a closed form expression for the unsaturated conductivity using an empirical expression for the water characteristic (water content - pressure head) relationship. The water content is expressed as:

$$S_e = \left(\frac{1}{1 + (\alpha h)^n} \right)^m \quad (4)$$

where α , n , and m are empirical constants determined from the shape of the water characteristic curve. Using $m = (1 - 1/n)$ an integral form of equation (2) may be simplified to the expression:

$$K_r = S_e^{1/2} \left[1 - (1 - S_e^{1/m})^m \right]^2 \quad (5)$$

The advantage of the van Genuchten approach over the approach of Campbell (1974) and others (Brooks and Corey 1964; Rogowski 1971) is the ability to fit data in the near saturation range (from air entry to zero pressure head), and to have a continuous function over the entire water content range of interest.

Another expression for unsaturated conductivity has been developed by Bresler, Russo and Miller (1978), hereafter called the BRM method. They developed an expression for the relative conductivity based on the wetting front advance into horizontal laboratory columns. Their expression is:

$$K_r = 0.27 m^4 (h_e/h)^{2.6} \quad (6)$$

where, m is the slope of a plot of distance to the wetting front versus the square root of time during horizontal infiltration, and h_e and h are the air entry and capillary pressure of the soil water.

This expression was derived for capillary pressures below (more negative than) the air entry pressure, and was based on the regression analysis of 7 different soils with widely varying texture and water flow characteristics (Bresler et al. 1978). When equation (5) is combined with the water characteristic expression suggested by Brooks and Corey (1964); i.e.,

$$S_e = \left(\frac{h_e}{h}\right)^\lambda \quad (7)$$

where λ for the 7 soils was determined by regression to be 0.36, the resulting expression is:

$$K_r = S_e^{7.2} \quad (8)$$

with $K_s = 0.27 \text{ m}^4$ (Bresler et al. 1978). A plot of this simple function against the K_r values of the soils used to develop the regression values yielded satisfactory results. An additional 12 soils from the literature (Mualem 1976) were also tested and equation (7) was found to fit the data in a satisfactory manner. The BRM analysis suggests that 7.2 may be a universal constant. Recently, Jones et al. (1979) tested equation (7) with data taken on Hanford Site burial ground soils. Their results indicated that the factor 0.27 m^4 did not satisfactorily predict the saturated water conductivity for tested soils. However, good agreement was obtained between the Millington-Quirk method (equation 2) and the Bresler, Russo, Miller (BRM) method (equation 8) when measured K_s values were used. For one soil, reasonable agreement was also found between field measured conductivity (Sisson and Gibbs 1979) and the BRM method (equation 8).

Another empirical relationship that has been used to a considerable extent relates the relative conductivity exponentially to the water content (Davidson et al. 1969, Nielsen et al. 1973, and Klute and Heermann 1978). This expression can be written as:

$$K_r = \exp [\hat{a} (\theta - \theta_s)] \quad (9)$$

where $\hat{a}$ is an empirical constant determined for each soil. Field data obtained in the wet range (draining soils) have been successfully fitted to this expression resulting in reasonable predictions of seasonal unsaturated water flux (Nielsen et al. 1973 and Simmons et al. 1979).

The particular choice of an unsaturated conductivity model for predicting water transport in burial ground soils is somewhat arbitrary. For arid region sites, the model should adequately predict the unsaturated flow characteristics in the dry range of water contents. Since virtually all of the models described have been tested and compared only in the wet range of soil water contents, it is still uncertain how well any of the models or empirical equations predict the hydraulic properties for relatively dry soils. The work reported in the following section on unsaturated water conductivity compares several of the available models with experimental data over the range of water contents from saturation to near air dry in a Hanford Site burial ground soil.

2.2 RADIONUCLIDE TRANSPORT

A recent evaluation of the sorption characteristics of radionuclides under unsaturated conditions (Gee and Campbell 1980) indicated that for the two Hanford sediments tested, mobile radionuclides like tritium, strontium, and cobalt (when complexed with EDTA) were sorbed similarly under unsaturated column or saturated batch test conditions. However, the unsaturated column tests proved valuable in assessing the transport of tritium, technetium-95m, iodine-125, and cobalt-60 (EDTA) through Hanford sediments since sorption characteristics could be measured with greater precision with the column tests than with the batch method.

The breakthrough curves (BTC) generated by Gee and Campbell (1980) were analyzed using the mobile-immobile water concepts of van Genuchten and Wierenga (1976a, and 1977a,b). This theory attempts to explain the asymmetric nature of a BTC by dividing the soil water into a mobile and an immobile phase. The longitudinal movement of solute is accomplished only by flow of the mobile phase water. In the mobile phase, convection, hydrodynamic dispersion, and sorption are the dominant processes of movement or retardation. In the immobile phase, sorption and molecular diffusion are the dominant mechanisms. This division of the water is easily understood for aggregated media where there is a distinct bi-modal pore size distribution. The small intra-aggregate pores contribute very little to the bulk flow of water as compared to the large inter-aggregate pores. When dealing with unconsolidated sands as discussed in Gee and Campbell (1980) and in this paper the division between mobile and immobile water becomes less obvious. There are no distinct aggregates; however, flow through sandy materials is particularly non uniform. This can be seen quite clearly while observing the "fingering" of a wetting front during infiltration even in small columns. This non uniformity is a result of the wide velocity distribution which exists in apparently uniform samples of sandy materials. The regions where velocities are extremely small in comparison with the bulk flow contain the "immobile" phase.

The transport of a sorbing radionuclide may be described using the above theory with two equations giving the concentration of solute in the mobile and immobile phases (C_m and C_{im}). There are four parameters; the Peclet number (P), mass transfer coefficient (α), and retardation factors (R and β) (van Genuchten and Wierenga 1976a). The Peclet number characterizes the dispersion within the column which indirectly is related to the velocity of flow and the size of the column, α describes the transfer of solute between the mobile and immobile phases, while R and β describe the sorption reactions. The significance of each of these parameters is discussed at length by van Genuchten and Wierenga (1976a). Independent measurement of these parameters is very difficult if not impossible, so a BTC curve fitting procedure as described by van Genuchten (1980) has been used.

The characterization of sorption reactions that remove radionuclides from solution in soils and sediments is a very active area of research. Among the diverse processes being studied are oxide and carbonate precipitation, precipitation of hydroxide compounds, lattice replacement, and ion exchange. In recent years the relative emphasis placed on ion exchange has decreased, however, it still is an important process for certain radionuclides. A fundamental property of ion exchange reactions is that they are dependent on the solution concentration of the exchanging ion as well as the ionic strength and composition of the bulk solution. Solute removal from soil exchange sites by mass action of a high salt solution following leaching by solution of a lower salt concentration has been called the snowplow effect by Starr and Parlange (1979). When the ion exchange capacity is low relative to the concentration of the displacing salt solution, and the relative affinity of the displacing ions for the exchanger is greater than those on the exchange sites, then simple mass action can result in near quantitative removal of ions initially present in the soil. This flushing of the soil column produces breakthrough curves (BTC) that are unusual in that when flushing occurs, the relative salt concentration, C/C_0 , in the effluent typically exceeds unity before dropping rapidly to zero. The magnitude of the BTC peak is dependent upon initial and final solute concentrations of the influent solutions.

The implication for shallow land burial sites is that errors can be made in predicting the movement of some radionuclides through sandy soils when only average salt concentrations are used in predictive equations rather than actual time dependent loading rates. Burial ground soils with relatively high concentrations of salt may, under increased rainfall, flush sorbed radionuclides from the soil exchange sites creating a much higher effluent concentration than predicted. Included in this report is verification of the "snowplow effect" for strontium-85. Conventional solute transport equations and those that include the mobile and immobile water concept (van Genuchten and Wierenga 1976b), are used to analyze the data.

The extension of these laboratory analyses to actual burial sites depends upon the ability of the parameters derived from small column, laboratory experiments to predict radionuclide transport in the field. The transfer from

laboratory to field involves a dramatic change of scale and also a much more complicated environment which needs to be described. Most laboratory studies are done on columns of 1-5 cm diameter and up to 50 cm in length. Burial grounds are generally several square hectares across and the depth of interest may be anywhere up to 100 meters. It has been well documented that the dispersion coefficient common to most transport equations is very dependent on experimental scale, and therefore, laboratory experiments may not be appropriate for the measurement of dispersion in field soils. It is not known if the sorption parameters and the mobile-immobile water parameters are also dependent on experimental scale. Field studies to examine this problem are currently being conducted at the Burial Waste Test Facility (BWTF), described by Phillips et al. (1979). As a preliminary step, laboratory experiments of an intermediate scale were also conducted and these data are presented in this report. A large column (0.5 m x 1.7 m) was constructed in the laboratory where the climate and solution flow rates could be controlled. The movement of tritiated water through the column was monitored and is presented along with analyses similar to the other column studies.

3.0 MATERIALS AND METHODS

3.1 SOILS

Rupert sand, a typic torripsamment, was used in the small and large column tests. Chemical and physical properties for this soil are given in a previous report (Gee and Campbell 1980). The Rupert sand is low in clay percentage (<4%) and in cation exchange capacity (5 meq/100 g). This burial ground soil has an alkaline pH (8.2) and sorbs radionuclides to a varying extent depending on the radionuclide and soil solution tested. For Rupert sand, species such as tritium, technetium-95, strontium-85, cesium-137, iodine-125, and cobalt-60 (complexed with EDTA) are relatively mobile in mixed salt solutions. Specific details on the adsorption of radionuclides for this soil are given by Gee and Campbell (1980).

3.2 SOLUTIONS

The solutions used in the tests were either tap water of low ionic strength, or two mixed salt solutions designated as low (L) and high (H) salt. The two solutions had the following compositions:

Low salt (L) - 0.15M NaNO_3 , 0.01M KNO_3 , 0.002M $\text{Ca}(\text{NO}_3)_2$

High salt (H) - 0.15M NaNO_3 , 0.15M KNO_3 , 0.01M $\text{Ca}(\text{NO}_3)_2$

3.3 CONDUCTIVITY MEASUREMENTS

Unsaturated conductivities for Rupert sand were measured both by steady state and transient methods. Steady flow measurements were made following procedures described by Klute (1972). The wet range conductivities were determined from the unsaturated flow columns used in the solute transport study (Gee and Campbell 1980). The dry soil conductivities were determined from a steady state evaporation experiment (Klute 1972). A large column (19 x 19 cm x 20 cm) was allowed to drain on a hanging water column then placed in a room at near constant relative humidity and allowed to dry at the surface for more than a 2 month period until a steady flow rate was measured from the calibrated supply of the hanging water column. After steady state was reached, the column

was sampled with depth and water content gradients calculated. The computed diffusivity was converted to conductivity by multiplying by the appropriate water capacity for the water contents of interest. The transient experiments in the dry range were run after the procedure of Rose (1968). This procedure involved packing drained soil uniformly in small columns (4.7 cm dia x 15 cm) and allowing the columns to evaporate into an atmosphere of controlled humidity for 10 days. The column was sectioned and a plot of water content versus $x/\sqrt{t}$ was used to compute the diffusivity (Rose 1968). Unsaturated hydraulic conductivities were also calculated by the procedures of Campbell (1974), Bresler, Russo, Miller (1978), Millington and Quirk (1961), van Genuchten (1980a), and Davidson et al. (1969).

3.4 RADIONUCLIDE TRANSPORT TESTS

3.4.1 Large Column Tests

Figure 1 shows the column configuration for the large column. The column was layered by placing a clean coarse sand at a density of 1.38 g/cm^3 at 116 cm to simulate textural stratification at a burial site. The total depth of the column was 176 cm and the Rupert sand was packed to a density of 1.74 g/cm^3 . Tap water was applied until drainage occurred from the bottom of the column. Then tritium (^3H) spiked water was added in daily pulses to the top of the column at the rate of 2.5 cm/day for a period of 16 days (beginning February 19, 1980), followed by tritium free water for an additional 15 days). Break-through curves were analyzed for tritium using standard liquid scintillation procedures, (Gee and Campbell 1980).

3.4.2 Snow Plow Effect

The "snow plow effect" was demonstrated using strontium-85 as a tracer applied to the soil column in a pulse of low salt solution and then subsequently flushed from the column using the high salt solution. The experimental equipment used for the tests is shown in Figure 2 and is similar to that used by van Genuchten and Wierenga (1977a,b). Influent solutions were applied at the upper end of soil columns packed in plexiglass cylinders of 5.4 cm inside diameter. Columns 27.5 cm long of Rupert sand were packed to a dry bulk density of 1.73 g/cm^3 . At the lower end of the vertically positioned column, a

porous plate for controlling vacuum and supporting the soil was attached. The column was connected to a vacuum chamber enclosing an automatic fraction collector. Figure 2 shows the apparatus used for measuring radionuclide transport in unsaturated soil.

The low salt solution containing a 50 nCi/ml spike of strontium-85 was pulsed through the column. After 4.3 pore volumes the strontium-85 was removed from the influent solution. The column was maintained at a moisture content of $0.17 \text{ cm}^3/\text{cm}^3$, and the flow rate was 6.8 cm/day. This run corresponds to the run dated 10/3/78 in Gee and Campbell (1980), Table 7. After 18.25 pore volumes of effluent were collected the relative concentration of strontium-85 (C/C_0) was <0.01 . The high salt solution containing no strontium-85 was then added to the column at a flow rate of 7.0 cm/day. The effluent was collected as before and the breakthrough curve determined.

FIGURE 1. Model of Large Laboratory Layered Soil Column

FIGURE 2. Apparatus for Measuring Radioruclide Movement in Unsaturated Soil Columns

3.4.3 Equations Used for Computer Simulations

The computer simulations discussed in this report fall into two categories: 1) curve fitting routines and 2) transport models. The fundamental equations of interest are the same for both types, however, the form is different. The transport models solve the equations in standard form while the curve fitting models solve the dimensionless form of the same equations. Both forms of the equation are presented here to help avoid confusion when the dimensionless parameter estimates obtained from the curve fitting routines are converted to the equivalent standard form so they can be used in the transport models. In this paper, there are two basic models being compared. The convective-dispersive model of solute transport is being compared to the mobile-immobile water model. In standard form the convective dispersive equation contains three parameters and is written:

$$\frac{\partial C}{\partial t} = \frac{1}{R} \left(D \frac{\partial^2 C}{\partial Z^2} - V_o \frac{\partial C}{\partial Z} \right) \quad (10)$$

where

C = solute concentration

t = time

D = dispersion coefficient

R = retardation factor $(1 + \rho K_d / \theta)$

Z = depth in soil

V_o = pore water velocity

In dimensionless form the same equation is written with only two dimensionless parameters as:

$$\frac{\partial C}{\partial t^*} = \frac{1}{R} \left(\frac{1}{P} \frac{\partial^2 C}{\partial Z^{*2}} - \frac{\partial C}{\partial Z^*} \right) \quad (11)$$

where

$P = \frac{V_o L}{D}$; the Peclet number

$$Z^* = \frac{Z}{L}$$

L = column length

$$t^* = \frac{V_o t}{L}$$

The mobile-immobile water model is really a system of two equations. In standard form there are two equations and six parameters. In dimensionless form there are only four parameters.

The standard form is:

$$\theta_m \frac{\partial C_m}{\partial t} + \theta_{im} \frac{\partial C_{im}}{\partial t} + f\rho \frac{\partial S_m}{\partial t} + (1-f)\rho \frac{\partial S_{im}}{\partial t} = \theta_m D \frac{\partial^2 C_m}{\partial Z^2} - V_m \theta_m \frac{\partial C_m}{\partial Z} \quad (12)$$

and

$$\theta_{im} \frac{\partial C_{im}}{\partial t} + (1-f) \frac{\partial S_{im}}{\partial t} = \alpha (C_m - C_{im}) \quad (13)$$

where

C_{im}, C_m = solute concentration in the immobile and mobile phases respectively

θ_{im}, θ_m = immobile and mobile water contents

V_m = mobile water velocity

f = fraction of sorption sites contacting mobile phase water

α = transport coefficient describing solute movement between mobile and immobile phases

ρ = soil bulk density

S_{im}, S_m = sorption in immobile and mobile phases.

Assuming $S = K_d C$ and using four dimensionless variables, equations 12 and 13 become:

$$\beta R \frac{\partial C_m}{\partial t^*} + (1 - \beta) R \frac{\partial C_{im}}{\partial t^*} = \frac{1}{P} \frac{\partial^2 C_{im}}{\partial Z^{*2}} - \frac{\partial C_{im}}{\partial Z^*} \quad (14)$$

and

$$(1 - \beta) R \frac{\partial C_{im}}{\partial t^*} = \bar{\alpha} (C_m - C_{im}) \quad (15)$$

where

$$\beta = \frac{\theta_m + \rho f K_d}{\theta + \rho K_d}$$

$$\bar{\alpha} = \frac{\alpha L}{V_m \theta_m}$$

$$P = \frac{V_m L}{D}$$

The computer simulations used to describe the snow plow effect were slightly more complicated. The hypothesis was that the sorption properties (i.e., the K_d value) was changing as a function of the bulk salt concentration. To test this the bulk salt concentration (G), had to be modeled as well as the solute concentration (C). When using the convective dispersion model (equation 10) to describe the solute transport, a second similar equation needed to be solved simultaneously that described the change in bulk salt concentration as the high salt solution moved through the column after it was introduced at 18.25 pore volumes.

This additional equation is:

$$\frac{\partial G}{\partial t} = D \frac{\partial^2 G}{\partial Z^2} - V_o \frac{\partial G}{\partial Z} \quad (16)$$

the retardation factor is set to one which is equivalent to setting $K_d = 0$. This is because the amount of the bulk solution ion sorbed is negligible in relation to the total salt concentration. The two equations are linked by describing the solute sorption (K_d), as a function of the bulk salt concentration (G). Exactly how this is done is described in Section 4.

A similar approach was used for the mobile-immobile water model. A second system of equations were written for the bulk salt concentration as:

$$\theta_m \frac{\partial G_m}{\partial t} + \theta_{im} \frac{\partial G_{im}}{\partial t} = \theta_m D \frac{\partial^2 G_m}{\partial Z^2} - V_m \theta_m \frac{\partial G_m}{\partial Z} \quad (17)$$

$$\theta_{im} \frac{\partial G_{im}}{\partial t} = \alpha (G_m - G_{im}) \quad (18)$$

4.0 RESULTS AND DISCUSSION

4.1 UNSATURATED CONDUCTIVITY MEASUREMENTS

The method of Campbell (1974) and the method of Bresler, Russo, and Miller (BRM), (1978) were compared against measured steady state values (Figure 3). The Campbell method is highly dependent upon the range over which the experimental coefficient b , (equation 3) is determined. In these tests, the slope (b) of the log-transformed pressure head versus water content curve was determined over the 10 to 100 cm matric potential range at the flexpoint of the water retention curve, as recommended by van Genuchten (1980). For soil bulk densities of 1.65 g/cm^3 and 1.80 g/cm^3 , the b values were 1.84 and 2.29, respectively. The two curves for the Campbell equation stay consistently within an order of magnitude and are in reasonable agreement with the experimentally measured values using steady state methods (Klute 1972). The Campbell equation is based on a capillary model which has no physical meaning when extrapolated to low water contents where vapor flow dominates. However, it does a remarkably good job in predicting the conductivity when the appropriate b is chosen. A value of 2 for b would give the best fit of the low water content data for Rupert sand. The BRM method (equation 8) under predicts hydraulic conductivity at very low water contents.

Figure 4 in addition to showing experimental steady state conductivities and calculated BRM conductivities compares the calculated hydraulic conductivities of Millington Quirk (1961) and Davidson et. al (1969) with the hydraulic conductivities measured under transient evaporation conditions of Rose (1968). The conductivities measured by the method of Rose (1968) are somewhat higher than the steady state values. The Millington and Quirk method under predicts the low water content conductivities for this soil. The exponential relationship of Davidson et al. (1969) was fit to obtain a soil parameter $\alpha = -32.2$ for Rupert sand so that equation 9 becomes:

$$K_r = \exp [-32.3 (\theta - \theta_s)] \quad (19)$$

FIGURE 3. Calculated and Measured Hydraulic Conductivities, Campbell and BRM Method

This equation (19) does a reasonable job in the wet range but over predicts the low range water conductivities by as much as three orders of magnitude.

The method of van Genuchten (1980a) was used to evaluate the hydraulic conductivity for the Rupert Sand used in the small and large column tests. Analytical expressions were developed for both the wetting and drying soil water characteristics. The analytical expressions for these curves are:

FIGURE 4. Calculated and Measured Hydraulic Conductivities, BRM, MQ and Davidson et al. Methods

$$\text{Drying curve: } \theta = 0.053 + \frac{0.267}{[1 + (0.027 h)^{2.95}]^{0.923}} \quad (20)$$

$$\text{Wetting curve: } \theta = 0.046 + \frac{0.274}{[1 + (0.074 h)^{1.88}]^{0.468}} \quad (21)$$

When these expressions were used to calculate the hydraulic conductivity, good agreement between calculated and experimental values was obtained in the wet range of soil water contents (Figure 5) with the experimental data falling on or within the calculated curves. The model failed to match the measured dry range soil water conductivities of the steady state or transient methods. The residual water contents, θ_r , obtained by curve fitting were 0.053 and 0.046

FIGURE 5. Hydraulic Conductivity Determined by the Method of van Genuchten from Wetting and Drying Water Characteristics of Rupert Sand Compared With Steady State Experimental Data

for the wetting and drying curves, respectively. van Genuchten (1980) has indicated that wet range conductivities are not significantly affected by the absolute values of the θ_r . On the other hand, the dry range values are significantly affected, since θ_r is defined as the water content at which hydraulic conductivity, $K = 0$.

The extensive work of Jackson (1964a,b,c, 1965) and Jackson et al. (1974) on water diffusion in dry soils indicates that the hydraulic conductivity curve can be extended to include the transport in the dry range only by accounting for water vapor diffusion. An effective conductivity which accounts for water vapor diffusion can be constructed using procedures of Fink and Jackson (1973), and Jackson (1964c) which more accurately predict the dry range hydraulic conductivity. The only input data required for this calculation is the water vapor isotherm for the soil in question. It therefore seems necessary to incorporate vapor diffusion characteristics into models where low water content data is of interest. For Hanford site burial grounds where sandy soil materials are dominant, the low water content range is of major interest. Field data show that virtually all burial ground soil profiles are drained to water contents of $<0.1 \text{ cm}^3/\text{cm}^3$. For well drained sandy sediments like Rupert Sand, typical drained water content values range from $0.05\text{--}0.07 \text{ cm}^3/\text{cm}^3$. Transport by isothermal vapor diffusion becomes increasingly more important in this range and the addition of a vapor flow term should improve the match between measured and calculated conductivities.

It is apparent from Figures 3, 4, and 5 that unsaturated hydraulic conductivities can be reasonably predicted in the wet range but care must be taken in extrapolating the equations to the low water content range. A complete analysis of vapor flow and coupled liquid and vapor flow would be required to assess the hydraulic conductivity in the dry region. Since well drained sediments at burial grounds of the Hanford site are normally drier than $0.1 \text{ cm}^3/\text{cm}^3$, additional effort is needed to evaluate the hydraulic conductivity relationship in this low water content range.

4.2 RADIONUCLIDE TRANSPORT MODELING

4.2.1 Curve Fitting Procedure

The parameter values used in the models were estimated by curve fitting solute breakthrough curves. The procedure used is a nonlinear least squares routine described by van Genuchten (1980b). The routine requires a first estimate of the parameters being fit. The curve fitting procedure was used twice on the data from the small column tritium and strontium experiments, in order to measure the reproducibility of the output. These runs are referred to in Table 7 of Gee and Campbell (1980). They are the strontium run of 10/27/78 and the tritium run of 11/27/78.

4.2.2 Small Column Experiments

Table 1 shows the parameter values obtained for both the two parameter convective-dispersive equation (equation 10), and the four parameter mobile-immobile water equations (equations 12 and 13). The parameter P , the Peclet number, theoretically depends only on the column length, soil moisture content, and the flow velocity. It should not depend on the solute. Table 1 shows that the four parameter fit produces an average P value for tritium of 106 while the average value for the strontium run is 29. The same column, soil, water content, and flow velocity, were used for each experiment. The Peclet numbers should be the same but they vary by more than a factor of 3. The two parameter model gives P values that vary by a factor of 6. This same observation was made by van Genuchten and Wierenga (1977b), when comparing tritium movement with movement of the pesticide 2,4,5-T. The second observation from analyzing these two data sets is that in some cases the two parameter equation seems to describe the data as well as the four parameter model. Figures 6 and 7 show the comparison of the two models together with the experimental data. For the strontium data, two distinct curves are produced, however in practical applications the two parameter model may prove as useful. The two curves produced from the tritium data were indistinguishable and are shown as one curve in Figure 7. The two parameter model can sometimes reproduce the curves generated by the four parameter model by adjusting the values P and R . Table 1 shows the two parameter model generally gives a much smaller P value and a

TABLE 1. Parameter Values Estimated by Curve Fitting Routine for Small and Large Column Experiments

<u>Isotope</u>	<u>Column</u>	<u>Fit No.</u>	<u>R</u>	<u>P</u>	<u>β</u>	<u>$\bar{\alpha}$</u>
^3H	Small	1	1.11	111	0.86	0.50
		2	1.11	102	0.86	0.42
		avg	1.11	106	0.86	0.46
^3H	Large	1	1.10	113	--	--
		1	1.17	22	0.76	0.36
		1	1.01	12	--	--
^{85}Sr	Small	1	8.4	28	0.72	0.47
		2	8.4	30	0.71	0.49
		avg	8.4	29	0.71	0.48
		1	7.2	19	--	--

slightly smaller R value. The tritium data shows something else, however. The R and P values are nearly identical between the two parameter and the four parameter models. It appears that setting $\bar{\alpha}$ and β equal to 0.50 and 0.86, respectively, does not alter the shape of the curve to any measurable degree. van Genuchten and Wierenga (1976a), Figure 4 show a significant affect on the shape of the curve by changing α from 0 to 0.15 (i.e., $\bar{\alpha}$ from 0 to 0.5). Apparently the large Peclet number (106) and small R value (1.11) of our experiment reduces the affect of $\bar{\alpha}$ and β on the shape of the curve. The Peclet number and R value used in the van Genuchten and Wierenga study were 38 and 2.6, respectively.

4.2.3 Large Column Experiment

The breakthrough curve generated for tritium from the large column experiment is shown in Figures 8 and 9. The data is more erratic than that of the small column experiments as is to be expected when large scale experiments are conducted. Any nonuniformity in the packing of the soil will affect the data and in this case, the presence of the gravel layer at the bottom of the column will have some effect on the flow. Table 2 shows the moisture content profile

FIGURE 6. ^{85}Sr Breakthrough Curve Using Small Column and High Salt Solution

during the course of the tritium experiment. It shows quite clearly that the gravel layer causes the water content directly above to increase to near saturation. The mixing in the saturated gravel layer will have some effect also. All these problems associated with large scale experiments are hard to describe in detail and equally hard to prevent.

Figure 8 shows the data points for both large column and small column tritium experiments. The data are similar with the large column showing

FIGURE 7. Tritium Breakthrough Curve Using Small Column and High Salt Solution

earlier solute breakthrough indicating more dispersion. This would be expected due to the increased column length. The tails of the breakthrough curves are difficult to compare because they are strongly influenced by the injection pluse width which was different for the large and small column. Figure 9 shows a comparison of a two parameter and a four parameter fit for the large column data. A two parameter fit corresponds to using the conventional convective dispersive equation (van Genuchten 1980b) while the four parameter fit comes from using the mobile-immobile water concepts (van Genuchten and Wierenga 1976a)

The two approaches do produce different predicted curves; however, it would be hard to say that one was better than the other. Table 1 shows the values of the parameter estimates found through the curve fitting routine. As mentioned above, it can be seen that using a two parameter equation requires a

FIGURE 8. Comparison of Tritium Breakthrough Curve Using Small and Large Column

much smaller Peclet number and a higher D . The Peclet number is defined as the water velocity times the column length divided by the dispersion coefficient. Generally a longer column produces a larger dispersion coefficient and therefore smaller Peclet number. Also, it is usually true that the four parameter model predicts a larger Peclet number and a smaller D than the two parameter model. This is because some of the asymmetry seen in the breakthrough curve can be accounted for in the $\bar{\alpha}$ and β terms, therefore requiring a smaller dispersion coefficient. The exception was seen to be the small column tritium experiment where the Peclet numbers were above 100. In the large column tritium experiment and the strontium experiments, where the Peclet numbers are approximately 10 to 30, the four parameter model predicts lower dispersion coefficients.

FIGURE 9. Comparison of Two and Four Parameter Models for Large Column Tritium Breakthrough Curve

One of the major problems in using laboratory data for field simulations is the variation of the dispersion coefficient (D) with both velocity and scale. The ratio D_L/D_S , the dispersion coefficient (D_L) of the large column experiment divided by the small column experiment (D_S), is equal to 31 for the two parameter model and 18 for the four parameter model.

The coefficients $\bar{\alpha}$ and β were consistent throughout all experiments. This may mean that for any soil these may be treated as constants regardless of column length or radionuclide. It has already been pointed out, from the small column tritium example, that these parameters appear to have little effect on the shape of the curve when the Peclet number exceeds 100. More work needs to be done with different tracers to characterize the variations of the parameters $\bar{\alpha}$ and β .

TABLE 2. Water Content Versus Depth for the Large Column During Tritium Experiments

<u>Depth, cm</u>	<u>Volumetric Water Content, cm³/cm³</u>
0	0.12
15.2	0.11
30.5	0.12
45.7	0.12
61.0	0.13
76.2	0.12
92.7	0.13
108.0	0.14
123.2	0.07
138.4	0.09
153.7	0.25

4.2.4 Snow Plow Effect

The breakthrough curve for this test is shown in Figures 10 and 11. The pore volumes start at 17. The tracer was stopped at approximately 4 pore volumes and the low salt solution stopped and high salt solution started at 18.25 pore volumes. The strontium was quickly released after the addition of the high salt solution. This is the result expected following the analysis of Starr and Parlange (1979). Also shown in Figures 10 and 11 are the results of the two and four parameter model simulations. The two parameter model used is described in detail by van Genuchten and Wierenga (1974) and the four parameter model by van Genuchten and Wierenga (1976b). The parameters used for the simulations were taken entirely from previous laboratory tests.

The dispersion coefficient (D) was determined using $P = 29$ for the four parameter model and $P = 19$ for the two parameter model. These values were taken from the high salt solution run shown in Table 1. The mobile water fraction (ϕ) was estimated using the reasoning of van Genuchten and Wierenga (1977a, 1977b).

FIGURE 10. Comparison of Two and Four Parameter Models for Snow Plow Effect

$$\phi = \beta R - f(R-1)$$

where

ϕ = mobile water fraction

f = fraction of sorption sites containing mobile water

β, R = curve fit retardation factors

The values of β and R were taken from the tritium run shown in Table 1, and used to calculate ϕ (equation 19). Letting f range from 0 to 1, results in a ϕ range of 0.85 to 0.95. An average value of 0.90 was used. The parameter f was estimated using β and R from the high salt solution run of strontium and the mobile water fraction from the tritium curve. This resulted in an estimate of $f = 0.69$. Three values for the K_d were used. The value of the low salt K_d was taken to be 6 from batch equilibrium studies (Gee and Campbell 1980), and high salt K_d was set at 0.745 for the four parameter model and 0.6 for the two parameter model. These values were obtained by using the R values from Table 1. The flow equations described in Section 3.4.3 were used to keep track of both the bulk soil solution salt concentration (G) as well as the

FIGURE 11. Sensitivity of Four Parameter Model to the Parameter f

radionuclide concentration (C). As the salt concentration in the column varied from low to high the K_d was varied linearly from 6 to 0.745 or 0.6. The results of these two simulation models are shown in Figures 10 and 11. Figure 10 shows the comparison between the two and four parameter approach. Figure 11 shows that by adjusting the value of f in the four parameter model a rather good fit may be obtained. There is no reason based on earlier experiments, however, to justify this adjustment. It was shown by Gee and Campbell (1980) that column K_d values are generally less than batch values. Using the batch K_d value of 1 for strontium in high salt would have moved both curves in Figure 10 far to the right. Conversely using a lower K_d value than 6 for the low salt would seem justifiable and would move both curves to the left and possibly improve the fit.

5.0 CONCLUSIONS

The hydraulic conductivity of unsaturated sediments is a difficult parameter to estimate. Several available models predict values consistent with each other within one order of magnitude. The agreement with experimental data is better in the wet range than the dry. This may be because most models do not explicitly include the contribution of water vapor flux to the total moisture flux. At low moisture contents this can produce significant errors. The main problem is shown to be establishing a correct value of the saturated hydraulic conductivity to use as a wet end match point and a residual water content as a match point on the dry end.

The radionuclide transport experiments described here seem to be described equally well by either the traditional convective-dispersive equation or the mobile-immobile water approach. The mobile-immobile water equations seem to be less sensitive to changes in experimental scale than the two parameter model, but the data suggesting this is from only one experiment. The snow plow effect was simulated reasonably well. The results of the snow plow experiment and model simulation are important for two reasons. First they confirm the hypothesis that changing the salt concentration of the bulk solution does affect the sorption properties of the soil. Secondly, this experiment is unique to this report because it attempted to use the curve fit parameter estimates previously obtained to simulate new experiments. Batch K_d values were used with column K_d values, and dispersion coefficients were used from other experiments. The results were a positive example of coupling both water flow and radionuclide sorption experiments to produce a simulation of radionuclide transport.

REFERENCES

- Bresler, E., D. Russo and R. D. Miller. 1978. "Rapid Estimate of Unsaturated Hydraulic Conductivity Function." Soil Sci. Soc. Am. J. 42:170-172.
- Brooks, R. H., and A. T. Corey. 1964. Hydraulic Properties of Porous Media. Colorado State University. Hydrology Paper 3, p. 27 Ft. Collins, Colorado.
- Burdine, N. T. 1953. "Relative Permeability Calculation From Size Distribution Data." Trans. AIME 198:71-78.
- Campbell, G. S. 1974. "A Simple Method for Determining Unsaturated Conductivity from Moisture Retention Data." Soil Sci. 117:311-314.
- Childs, E. C., and N. Collis-George. 1950. "The Permeability of Porous Media." Proc. Roy. Soc. Ser A 201:392-405.
- Davidson, J. M., L. R. Stone, D. R. Nielsen and M. E. Larue. 1969. "Field Measurement and Use of Soil-Water Properties." Water Resources Res. 5:1312-1321.
- Fink, D. H., and R. D. Jackson. 1973. "An Equation for Describing Water Vapor Adsorption Isotherms of Soils." Soil Sci. 116:256-261.
- Gee, G. W., and A. C. Campbell. 1980. Monitoring and Physical Characterization of Unsaturated Zone Transport-Laboratory Analysis. PNL-3304. Pacific Northwest Laboratory Richland, WA 99352.
- Gee, G. W., and C. S. Simmons. 1979. Characterization of the Hanford 300 Area Burial Grounds, Task III - Fluid Transport and Modeling. PNL-2921, Pacific Northwest Laboratories, Richland, WA 99352
- Green, R. E., and J. C. Corey. 1971. "Calculation of Hydraulic Conductivity: A Further Evaluation of Some Predictive Methods." Soil Sci. Soc. Am. Proc. 35:3-8.
- Jackson, R. D. 1964a. "Water Vapor Diffusion in Relatively Dry Soil: I. Theoretical Considerations and Sorption Experiments." Soil Sci. Soc. Am. Proc. 28:172-176.
- Jackson, R. D. 1964b. "Water Vapor Diffusion in Relatively Dry Soil: II. Desorption Experiments." Soil Sci. Soc. Am. Proc. 28:464-466.
- Jackson, R. D. 1964c. "Water Vapor Diffusion in Relatively Dry Soil: III. Steady-State Experiments." Soil Sci. Soc. Am. Proc. 28:466-470.
- Jackson, R. D. 1965. "Water Vapor Diffusion in Relatively Dry Soil: IV. Temperature and Pressure Effects on Sorption Diffusion Coefficients." Soil Sci. Soc. Am. Proc. 28:466-470.

- Jackson, R. D., B. A. Kimball, R. J. Reginato and F. S. Nakayama. 1974. "Diurnal Soil-Water Evaporation: Time-Depth-Flux Patterns." Soil Sci. Soc. Am. Proc. 37:505-507.
- Jones, T. L. 1978. Sediment Moisture Relations: Lysimeter Project 1976-1977 Water Year. RHO-ST-15, Rockwell Hanford Operations, Richland, WA 99352.
- Jones, T. L., W. A. Jordan and G. W. Gee. 1979. A Test of the Bresler-Russo-Miller Method for Unsaturated Conductivities. RHO-SA-125, Rockwell Hanford Operations, Richland, WA 99352.
- Klute, A. 1972. "The Determination of the Hydraulic Conductivity and Diffusivity of Unsaturated Soils." Soil Sci. 113:264-276.
- Klute, A., and D. F. Heermann. 1978. Water Movement in Uranium Mill Tailings. Tech Note ORPILV-78-8 p. 81. United States Environmental Protection Agency.
- Millington, R. J., and J. P. Quirk. 1961. "Permeability of Porous and Solids." Trans. Faraday Soc. 57:1200-1206.
- Mualem, Y. 1976. "A New Model for Predicting the Hydraulic Conductivity of Unsaturated Porous Media." Water Resour. Res. 12:513-522.
- Nielsen, D. R., J. W. Biggar and K. T. Erh. 1973. "Spatial Variability of Field-Measured Soil-Water Properties." Hilgardia 42:215-260.
- Phillips, S. J., A. C. Campbell, M. D. Campbell, G. W. Gee, H. H. Hooper and K. O. Schwarzmiller. 1979. A Field Test Facility for Monitoring Water/Radionuclide Transport Through Partially Saturated Geologic Media: Design, Construction, and Preliminary Description. PNL-3226, Pacific Northwest Laboratory, Richland, WA 99352.
- Reichardt, K., D. R. Nielsen and J. W. Biggar. 1972. "Scaling of Horizontal Infiltration Into Homogeneous Soils." Soil Sci. Soc. Am. Proc. 36:241-245.
- Rogowski, A. S. 1971. "Watershed Physics: Model of the Soil Moisture Characteristic." Water Resources Res. 7:1575-1582.
- Rose, D. A. 1968. "Water Movement in Porous Materials III. Evaporation of Water from Soil." Brit. J. Appl. Phys. 1:1779-1790.
- Sisson, J. B., and A. G. Gibbs. 1979. Application of Hydraulic Conductivity Scaling to Infiltration-Drainage Field Tests. RHO-SA-121 Rockwell Hanford Operations Richland, Washington.
- Starr, J. L., and J. Y. Parlange. 1979. "Dispersion in Soil Columns: The Snowplow Effect." Soil Sci. Soc. Am. J. 43:448-450.

- van Genuchten, M. Th., and P. J. Wierenga. 1974. "Simulation of One-Dimensional Solute Transfer in Porous Media." Agricultural Experiment Station Bulletin 628. New Mexico State University, Las Cruces, New Mexico.
- van Genuchten, M. Th., and P. J. Wierenga. 1976a. "Mass Transfer Studies in Sorbing Porous Media. I. Analytical Solutions." Soil Sci. Soc. Am. J. 40:473-480.
- van Genuchten, M. Th., and P. J. Wierenga. 1976b. "Numerical Solution for Convective Dispersion with Intra-Aggregate Diffusion and Non-Linear Adsorption." In System Simulation in Water Resources G. C. Vansteenkiste, ed. North Holland Publishing Company, Oxford, New York.
- van Genuchten, M. Th., and P. J. Wierenga. 1977a. "Mass Transfer Studies in Sorbing Porous Media II. Experimental Evaluation with Tritium ($^3\text{H}_2\text{O}$)."
Soil Sci. Soc. Am. J. 41:272-278.
- van Genuchten, M. Th., and P. J. Wierenga. 1977b. "Mass Transfer Studies in Sorbing Porous Media III. Experimental Evaluation with 2, 4, 5-T" Soil Sci. Soc. Am. J. 41:278-285.
- van Genuchten, M. Th. 1980a. "A Closed Form Equation for Predicting the Hydraulic Conductivity of Unsaturated Soils." Soil Sci. Soc. Am. J. 44:892-898.
- van Genuchten, M. Th. 1980b. Determining Transport Parameters from Solute Displacement Experiments. U.S. Salinity Laboratory Research Report No. 118. USDA/SEA, Riverside, CA.

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