A COMPARISON OF POSSIBLE MODEL AND PARAMETER ERRORS ASSOCIATED WITH SOLUTE RETARDATION IN GROUNDWATER TRANSPORT

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ABSTRACT

Groundwater solute transport models were developed which compared linear to one type of non-linear adsorption. Most traditional transport simulations rely upon linear adsorption in the form of solute retardance. While generally held to be appropriate at low solute concentrations, linear approximations are inadequate for some other applications. The subject work addressed this model uncertainty by comparing simulations completed assuming that Freundlich adsorption was appropriate with those developed under linear assumptions. A non-linear retardance equation was derived. Solute concentration is an equation variable rather than a constant and its estimation becomes critical in accurately defining transport times and distances. Errors associated with the selection of linear adsorption when non-linear retardation more appropriatedly applied ranged to 18.5 percent at low solute concentrations with variations in the Freundlich exponent. Error values approaching 13 percent were associated with solute concentration variation, while uncertainty in selecting the partition coefficient accounted for less than 7 percent.

Due to an increasing awareness of groundwater problems associated with contamination, a general lack of applicable monitoring data and the increasing availability of computers and software, pollution events are routinely modeled by mathematical simulation. Models are used by both industry and governmental agencies involved in groundwater management aids in decision making. This ever increasing reliance on models has raised concerns as to the errors associated with their uses [1]. Inability to simulate actual conditions found in nature may cast doubts on those environmental decisions resulting from extensive use of

simulation. This point was discussed recently in a National Research Council study which recommended that extreme care be exercised in deciding groundwater management controversies when the primary weight of evidence was based solely upon transport modeling [2]. Numerous factors affect the accuracy and precision of model simulations; some of the most important are the description of transport mechanisms, proper choice of model codes and the accurate utilization of input parameters.

Two types of uncertainties may distort the results of these simulations. First, model uncertainty can result from the improper selection of a model formulation, and second, parameter uncertainty can occur when a potentially wide range of often difficult-to-acquire input data are used within the chosen code. The reliability of the model simulations can be greatly affected by either or both of these uncertainties. This work evaluated the simultaneous impact of both types of uncertainties when transport of organic solutes through sorbing media occurred. Specifically, the effects of choosing alternative retardance algorithms and attendant model parameters were determined. A brief overview of adsorption in groundwater systems with emphasis upon possible sources of uncertainty follows.

In addition to adsorption, organic chemicals can undergo a variety of transport and transformation reactions upon introduction into an aquifer. Advection, dispersion, hydrolysis, oxidation-reduction, volatilization, and biodegradation, for example, may affect the ultimate contaminant concentration. Each of these offer potential sources of error either from the selection of functional forms describing these processes or in the application of specific parameters to each. Errors associated with these, while potentially important, were not considered in this effort.

A contaminant is relatively immobile and will not be transported from a source if the chemical is strongly adsorbed to the solid matrix, which may display a high adsorption capacity. If the chemical is weakly adsorbed, or the solid matrix displays a low adsorption capacity, it can be leached from the source and be transported long distances thus contaminating the aquifer through which it flows [3].

Adsorption is incorporated into the transport simulation equation by terms representing the interactions between the aquifer solids (adsorbent) and the solute (adsorbate). The actual partitioning between the solid and liquid phases by a chemical compound may be described by a particular functional form called an adsorption isotherm. The isotherm is dependent upon the interactions between the two phases, the properties of adsorbate and the chemical concentrations found in the solid and liquid phases. A general partitioning between these phases can be written using the Freundlich equation:

$$q = (K_f) \, C^{1/n} \tag{1}$$

where q is the adsorbed or solid phase concentration, K_f is the Freundlich adsorption constant, $1/n$ is an experimentally derived exponent, and C is the concentration of the adsorbate in the solution [4-6]. Although the Freundlich isotherm

equation was developed empirically, a theory of adsorption that leads to its application was later presented where the constant, K_f, was related to the capacity of the adsorbent for the adsorbate while $1/n$ was a function of the strength of the adsorption bond [7]. For fixed values of C and $1/n$, the greater the value of K_f, the larger the adsorptive capacity q. For fixed values of K_f and C, the smaller the value of $1/n$, the stronger the adsorption bond [8]. As $1/n$ becomes very small, the adsorption capacity tends to be independent of C and the isotherm plot approaches the horizontal; the value of q then is essentially constant and the isotherm is called irreversible. If the value of $1/n$ is large, the adsorption bond becomes weak and the value of q changes markedly with small changes in C.

These later two conditions describe saturation based adsorption more commonly typified by the monolayer Langmuir isotherm. For this reason, the Freundlich equation is occasionally held to be based on monomolecular adsorption at heterogeneous sites [9]. For this reason, the Freundlich equation is often considered to be appropriate for soil/aquifer systems which have highly variable adsorptive affinities [8]. Unlike the more commonly utilized linear applications, the Freundlich isotherm equation does not apply to all values of solute concentration, C. As C increases, q also increases until the adsorptive capacity approaches saturation. At saturation, q is constant and independent of further increases in C and the Freundlich isotherm equation is no longer applicable. Caution must be exercised when extending the Freundlich isotherm equation over concentration ranges that have not been previously tested.

For a variety of chemicals and under certain circumstances, $1/n$ is equal to unity and Equation (1) reduces to:

$$q = (K_d) \, C \tag{2}$$

This linear adsorption isotherm is valid only under equilibrium conditions and only if adsorption is proportional to solute concentration [10]. Equilibrium conditions prevail when the reactions that cause the partitioning are reversible and proceed quickly when compared to the aquifer flow velocity. As noted previously, these conditions are met on only a few occasions when either chemical or adsorbent specific properties allow.

Adsorbates are held on the surface of adsorbents by various types of chemical forces such as hydrogen bonds, dipole-dipole interactions and van der Waals forces [11, 12]. If the reaction is reversible, adsorbate molecules accumulate on the adsorbent surfaces until the rate of adsorption equals the rate of desorption. When this condition is met equilibrium has been reached and no further net adsorbate accumulation on the solid phase will occur. Many organic compounds are thought to follow a linear adsorption isotherm at low concentrations [13, 14, 15]. It can be seen from Equation (2), when $K_d = 0$, no adsorption on the adsorbent will occur and the contaminant will flow through the aquifer at the same velocity as the water. If $K_d > 0$ the contaminant will flow through the

aquifer at a slower mean velocity than the water thus displaying the characteristic of retardance [16].

Kd is conceptually defined as the ratio of concentration in the solid phase to that in the liquid phase:

$$K_d = \frac{C_s}{C_1} \tag{3}$$

C_s (mass of adsorbate on the adsorbent per unit mass of adsorbent) can be expressed in terms of mg/Kg and C_1 (concentration of adsorbate in solution) can be expressed in terms of mg/L. Therefore K_d carries the units of ml/g.

Values of K_d for an organic compound will vary depending upon the adsorptive properties of the adsorbent as well as the amount of organic matter present in the solid phase. K_d, in Equation (3), has been approximated by the product of the soil organic carbon partition coefficient, k_{oc}, and the soil organic carbon matter content, f_{oc} [16] as presented in Equation (4). It has been shown, however, that adsorption to mineral surfaces may exceed that to soil organic matter at low adsorbent levels [17] and that different soil organic components will have varying adsorptive capacities [18, 19].

$$K_d = (k_{oc})\,(f_{oc}) \tag{4}$$

A mass balance equation for solute transport under the influences of advection, dispersion and adsorption can be written as [20]:

$$\frac{\partial C}{\partial t} = -U\frac{\partial C}{\partial x} + D\frac{\partial^2 C}{\partial x^2} - \frac{\rho b}{\varepsilon}\frac{\partial q}{\partial t} \tag{5}$$

where:
  C = Liquid phase substrate concentration
  t = Time
  U = Interstitial groundwater velocity
  x = Distance
  D = Dispersion coefficient for substrate
  ρb = Bulk density of the adsorptive medium
  ε = Porosity of adsorptive medium
  q = Solid phase substrate concentration

The last term in Equation (5) is the expression for adsorption. If the distribution of adsorbate between the liquid and solid phases can be described by the linear adsorption isotherm given in Equation (2), the adsorbate concentration on the solid phase is proportional to the concentration in the liquid phase. By applying the differential train rule:

$$\frac{\partial q}{\partial t} = \frac{\partial q}{\partial C}\frac{\partial C}{\partial t} \tag{6}$$

and combining with the differential of Equation (2)

$$\frac{\partial q}{\partial C} = K_d \tag{7}$$

$$\frac{\partial q}{\partial t} = (Kd)\frac{\partial C}{\partial t} \tag{8}$$

and substituting Equation (8) into Equation (5) and recombining

$$\frac{\partial C}{\partial t} = -U\frac{\partial C}{\partial x} + D\frac{\partial^2 C}{\partial x^2} - \frac{\rho b K_d}{\varepsilon}\frac{\partial C}{\partial t} \tag{9}$$

an expression for retardance under linear adsorption conditions, R in Equation (10), can be derived. This form is often used interchangeably with K_d for a given bulk density and porosity of the adsorbent.

$$R = \left(1 + \frac{\rho b K_d}{\varepsilon}\right) \tag{10}$$

For some complex organic compounds or at elevated concentrations, Hamaker and Thompson [21] have shown that $1/n$ is not equal to unity thus indicating nonlinear adsorption. Using the above method, a retardance factor for nonlinear adsorption behavior based upon Freundlich principles can also be derived. By applying the differential train rule to Equation (6) and combining with the differential of Equation (1)

$$\frac{\partial q}{\partial C} = \frac{K_f C^{1/n-1}}{n} \tag{11}$$

$$\frac{\partial q}{\partial t} = \frac{K_f C^{1/n-1}}{n}\frac{\partial C}{\partial t} \tag{12}$$

and substituting Equation (12) into Equation (5) and recombining:

$$\frac{\partial C}{\partial t} = -U\frac{\partial C}{\partial x} + D\frac{\partial^2 C}{\partial x^2} - \frac{\rho b K_f C^{1/n-1}}{\varepsilon n}\frac{\partial C}{\partial t}$$

$$\frac{\partial C}{\partial t}\left(1 + \frac{\rho b K_f C^{1/N-1}}{\varepsilon n}\right) = -U\frac{\partial C}{\partial x} + D\frac{\partial^2 C}{\partial x^2} \tag{13}$$

an expression for nonlinear Freundlich retardance, R', is produced:

$$R' = \left(1 + \frac{\rho b K_f C^{1/n-1}}{\varepsilon n}\right) \tag{14}$$

This nonlinear retardance is dependent upon concentration, C, which varies spatially and temporally along the flow paths within an aquifer while linear retardance, R, being independent of concentration, remains constant when used in model formulations. Nonlinear retardance, however, will continue to vary with C, as well as with $1/n$ and K_f, until saturation is obtained.

A well-understood example illustrating the importance of understanding nonlinear adsorptive behavior is the application of pesticides to agricultural fields. If the system is truly linear, as is depicted in the most commonly available agrichemical transport codes, the relative amount of pesticide leaching through the upper soil horizons will remain constant and proportional to that applied. The proportionality constant would be equal K_d. In reality, as the upper soil adsorptive sites become occupied with adsorbed solute, a greater percentage of the applied pesticide will migrate to the aquifer. A saturation level of sorbed pesticide is reached allowing an increased percentage of chemical to leach to greater depths. This condition is not described by linear formulations. That is, model uncertainty exists when linear retardance is used to describe these nonlinear conditions. Further, assuming nonlinear adsorption as described by the Freundlich equation is a correct approximation of mass transfer within the aquifer, uncertainty in the selection of K_f, C or $1/n$ can introduce additional errors further reducing the applicability of the simulation.

EXPERIMENTAL RESEARCH STRUCTURE

This article compares the variance error which resulted when a traditional linear equilibrium approach was applied to simulate transport where Freundlich adsorption more appropriately described the process. Further, the variance and mean errors resulting from the selection of Freundlich parameters, K_f, $1/n$ and C over a wide range of values are identified.

To compare the results using linear adsorption to simulate Freundlich nonlinear adsorption the linear adsorption constant K_d was held equivalent to the corresponding Freundlich K_f. From Equations (1) and (2), it can be shown that when $1/n$ is set equal to unity the nonlinear model describes linear adsorption. Linear retardance from one to ten was selected to approximate a range of appropriate transportation conditions [22]. This range defines mean solute transport times from that equivalent to mean hydraulic detention to ten times this value. From Equation (10), a corresponding value of K_d was calculated for each linear retardance value and for fixed levels of porosity (40%) and soil bulk density (1.2 gm/cm^3). These are presented in Table 1.

From the last term in Equation (11), the expression for nonlinear adsorption in the mass balance for solute transport, it is clear that nonlinear adsorption will vary with values of K_f, $1/n$ and C. Therefore for each value of K_d in Table 1 a family of breakthrough curves was generated by varying values for both

Table 1. Values for Linear
Retardance and K_d

Linear Retardance	K_d (cm^3/gm)
1	0.000
2	0.333
3	0.666
4	1.000
5	1.333
6	1.666
7	2.000
8	2.333
9	2.666
10	3.000

$1/n$ and C. Literature values were used in this study to represent commonly occurring parameter ranges allowing direct comparisons to other efforts [23, 24].

The concentration of adsorbate in solution, C, was modeled using a constant boundary input concentration ranging from 0.5 ppm, a common regulatory limit for many organic chemicals, to a value over five orders of magnitude greater. This approach provided breakthrough curves for contaminant concentration, from the lower limit of detection to saturation, where no additional contaminant would sorb to the adsorbent. These values were selected more for their properties relative to defining a linear-nonlinear saturation curve rather than for similarities to known specific chemicals or conditions. In this manner, the analyses were felt to be more descriptive of a variety of potential modeling conditions. A recap of the range of values used in this effort for $1/n$ and C are presented in Table 2 while Table 3 summarizes the fixed data employed.

Following the work of Chen and McTernan [25], the use of dimensionless time within the aquifer volume allowed comparisons between varying conditions inherent in other simulations. That is, dimensionless time as defined by retardance or relative travel time was used to standardize results so that comparisons between simulations could be made.

MODEL AND ANALYSIS DESCRIPTION

OklahomaState University's Multi-Substrate, Multi-Option Groundwater Transport Model (MMGTM) was used to generate the breakthrough curves analyzed in this effort [26]. In addition to the normal advective-dispersive

Table 2. Values for 1/n and C

1/n	C (mg/l)
0.133	0.5
0.333	5.0
0.533	50.0
0.733	100.0
0.933	450.0
1.000	900.0
	1300.0

Table 3. Constant Parameter Values

Parameter	Value
Interstitial velocity	0.2 m/day
Dispersion coefficient	0.4 m^2/day
Aquifer bulk density	1.2 gm/cm^3
Aquifer porosity	40.0%
Nodal distance	5.0 m

transport, MMGTM can concurrently simulate numerous adsorption and bio-logical decay functions. Linear or nonlinear adsorption, represented by Freundlich and Langmuir isotherms, under either equilibrium or non-equilibrium conditions can be selected to better describe the true adsorptive properties. The model, as used in this study, defined differences between linear and non-linear Freundlich adsorption at equilibrium. That is, the objective of this effort was to identify the amount of error introduced in a simulation based upon linear equilibrium adsorp-tion when the conditions were more appropriately described by one specific nonlinear formulation.

Variance error, defined as the difference between the breakthrough concentra-tions, normalized to the input boundary concentration, resulting from linear and nonlinear adsorption simulations was initially employed to make these types of comparisons. These determinations were made in accordance with Equation (15) where breakthrough concentrations resulting from each of the nonlinear simula-tions were subtracted sequentially from the linear breakthrough concentrations determined at appropriate dimensionless times.

$$E_v = (C_1 - C_{nl}) \tag{15}$$

where:
E_v = Variance error
C_l = Normalized concentration for linear adsorption
C_{nl} = Normalized concentration for nonlinear adsorption

The results obtained from the analyses generated a series of error curves over the range of nonlinearities investigated at the dimensionless detention times selected. When combined, these curves provided an envelop of predicted errors for the conditions inherent in each simulation exercise. These determinations were not corrected for the dimensionless time increments employed nor for the total summing period. As such they represented a running error sum over identical time scales for each set of simulations. They have utility when used to identify trends in expected errors associated with model and parameter uncertainties. The general magnitude and the relative shapes of these curves illustrate the range of possible errors associated with the model and parameter selections employed in this effort. More precise determinations of expected errors, however, are best completed by mean error comparisons which follow.

Mean error, defined as the sum of the square of the difference between the normalized concentrations for linear and nonlinear adsorption divided by the total number of dimensionless time increments was determined for each of the linear-nonlinear comparisons previously described by Equation (16).

$$E_m = (\Sigma(C_1 - C_{nl})^2 / N\Delta t)^{1/2} \tag{16}$$

where:
E_m = Mean error
C_l = Normalized concentration for linear adsorption
C_{nl} = Normalized concentration for nonlinear adsorption
$N\Delta t$ = Number of time increment

RESULTS

Figures 1, 2, and 3 present example variance error plots for boundary solute concentrations of 0.5, 100, and 450 mg/l respectively. Similar figures for solute levels of 5, 50, 900, and 1300 mg/l were also completed but are excluded from further discussion as they followed closely the trends established in these Figures. Figure 1 shows that strong overprediction (i.e., positive variance error) would be expected from the linear model at low solute concentration when the nonlinear assumption was more appropriate. Figures 2 and 3, however, suggest that the corresponding errors could be either positive (linear overpredicted) or negative (under predicted) as boundary solute concentration increased. Additional solute concentration increases beyond 450 mg/l resulted in no significant alterations to the errors traced in Figure 3.
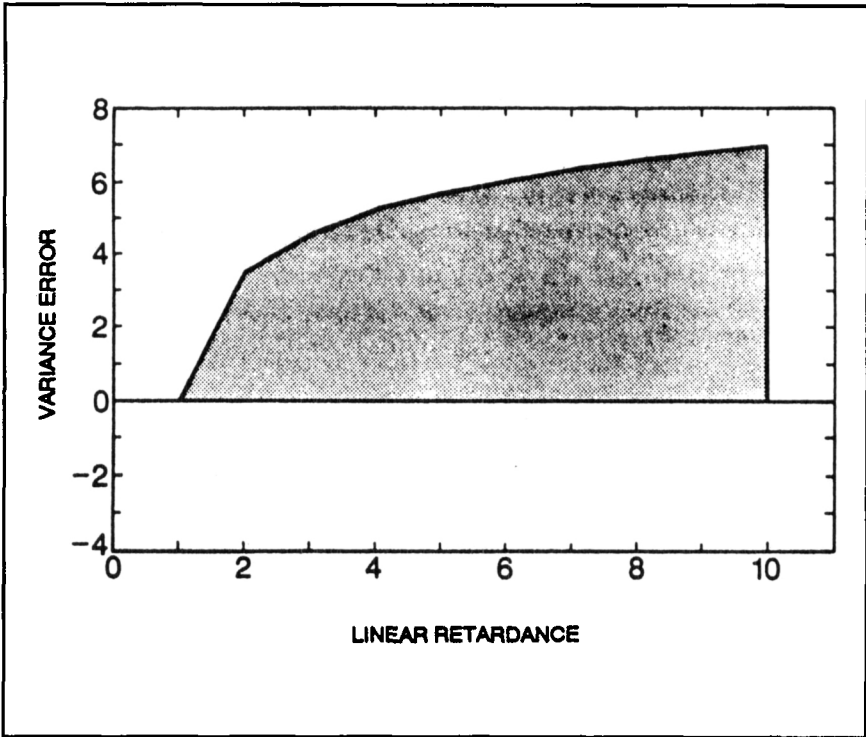
Figure 1. Variance error versus linear retardance at solute
concentration of 0.5 mg/l.

Figure 4 presents example mean error plotted against linear retardance in
the aquifer at varying 1/n values for constant solute concentrations (C) of 5 and
900 mg/L respectively. It can be seen that the magnitude of the mean error
increased with adsorbate boundary concentration but, as expected, decreased to
zero at a linear retardance of unity. For a specific linear retardance value and a
continuous adsorbate boundary concentration, a range of mean error values can
be obtained from Figures 5, 6, and 7, where solute concentrations of 0.5, 100, and
450 mg/L respectively were employed. The specific mean error to be expected
may be any value within this range, depending upon the magnitude of the
Freundlich exponent, 1/n. Decidedly nonlinear behavior at corresponding retar-
dance or dimensionless detention times was observed. The affects of uncertainty in
identifying precise values of Kd, 1/n, and C over an extensive range are illustrated
in these figures. While the mean error generally increased with increasing differen-
ces retardance, a range of less than 7 percent was observed with the greatest
differences occurring as the boundary concentration increased. Further, mean error
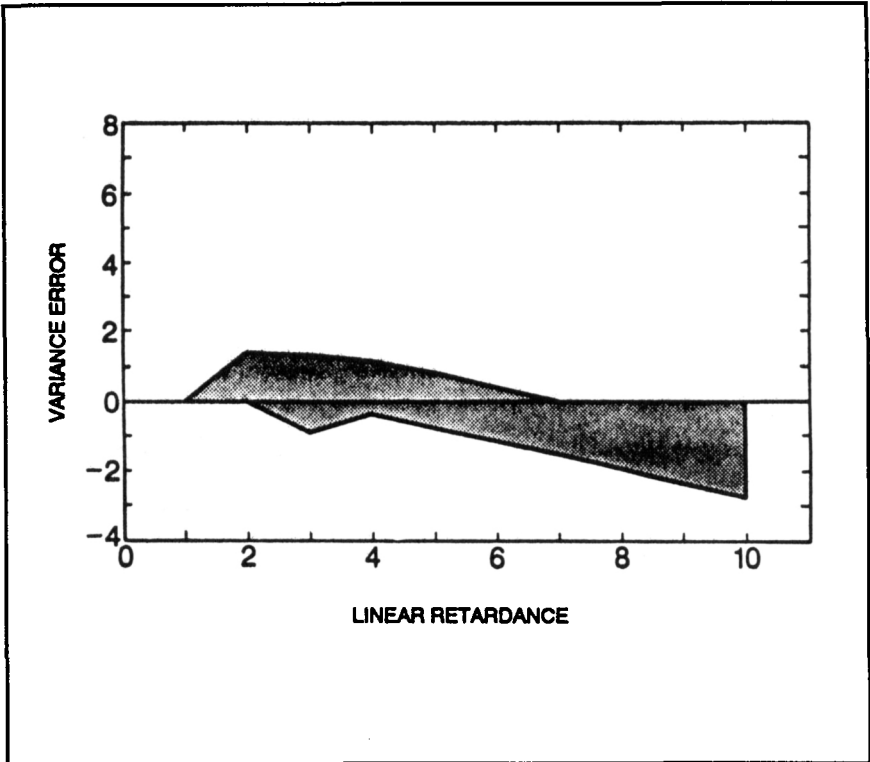resulting from the uncertainty in selecting a boundary solute concentration (C) was

Figure 2. Variance error versus linear retardance at solute
concentration of 100 mg/l.

less than 13 percent over the linear retardance range modeled and decreased as the boundary concentration increased. The mean error achieved a maximum value of 18.5 percent at a constant boundary adsorbate concentration of 0.5 mg/L, when solute detention times were six, eight or ten times the mean liquid detention.

The mean error was only slightly affected by an increase in boundary adsorbate concentration once the value exceeded 1 mg/L. The major factor, however, affecting mean error appeared to be the Freundlich exponent, 1/n. Virtually all of the measured variation was attributable to this parameter. Further, interactions between the exponent and the boundary concentration resulted in the majority of the nonlinear responses.

SUMMARY AND CONCLUSIONS

This research addressed the uncertainty which could result with the selection of linear equilibrium adsorption when specific nonlinear conditions applied. For a
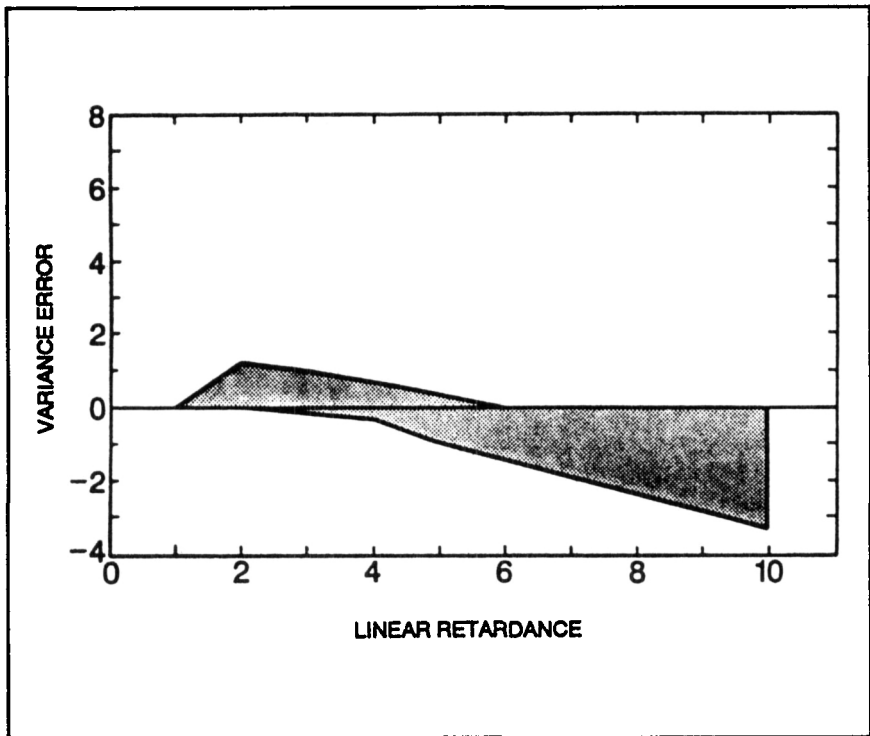
Figure 3. Variance error versus linear retardance at solute
concentration of 450 mg/l.

continuous boundary input condition, the selection of a linear adsorption model
was found to over-predict the solute concentration simulated by the nonlinear
model at low boundary concentration values and to be under-predictive at higher
boundary input values. In comparing corresponding curves it was noted that at
solute concentrations approaching saturation, the breakthrough concentrations
predicted from the linear models were less than those from the nonlinear
approach. This underscored the potential problems present when using an under-
predictive linear model at high solute concentrations. The time required for
contaminant breakthrough decreased as the boundary adsorbate concentration was
increased. This was expected due to the effect of saturation on the adsorbent sites
available for adsorption.

As shown in Figures 4 through 7, the mean error resulting from the uncertainty
in selecting K_d was less than 7 percent over the range of linear retardance modeled
and displayed a wider range as the boundary concentration increased. Mean error
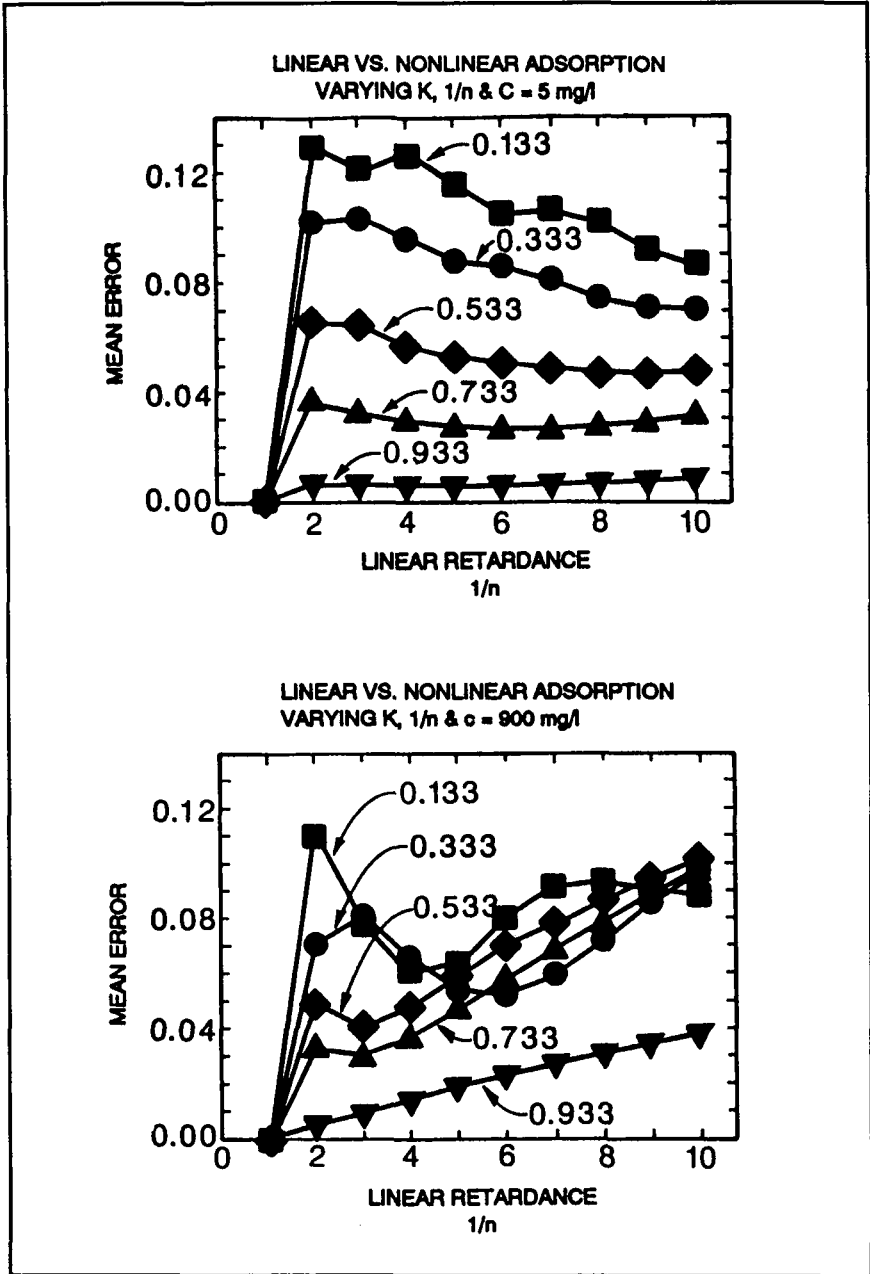resulting from the uncertainty in selecting a boundary solute concentration (C)

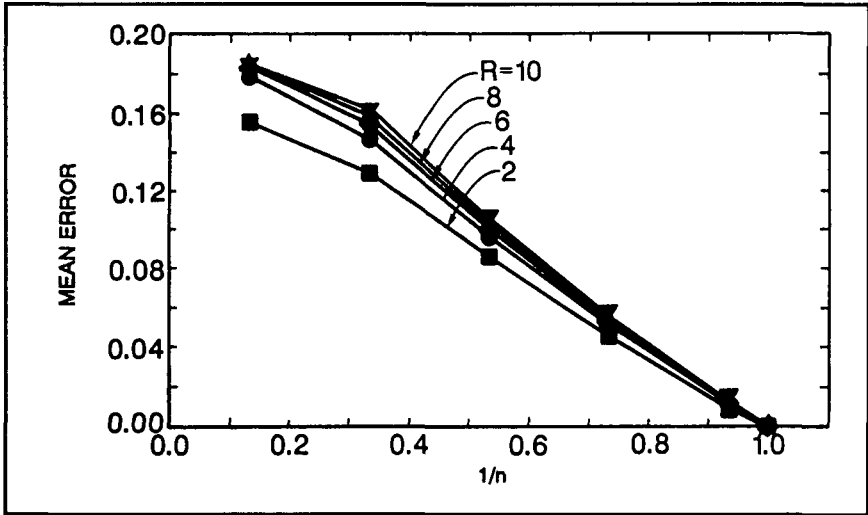Figure 4. Mean error versus linear retardance at solute concentrations of 5 and 900 mg/l.

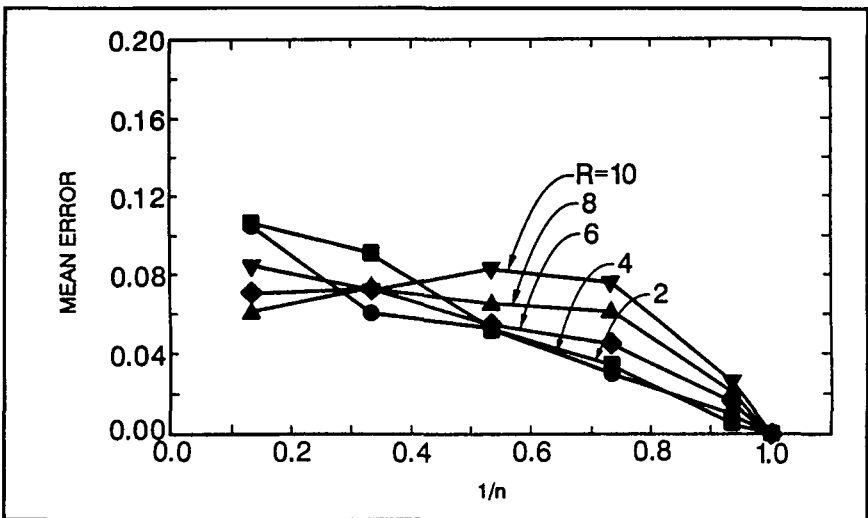Figure 5. Mean error versus 1/n at solute concentration
of 0.5 mg/l.

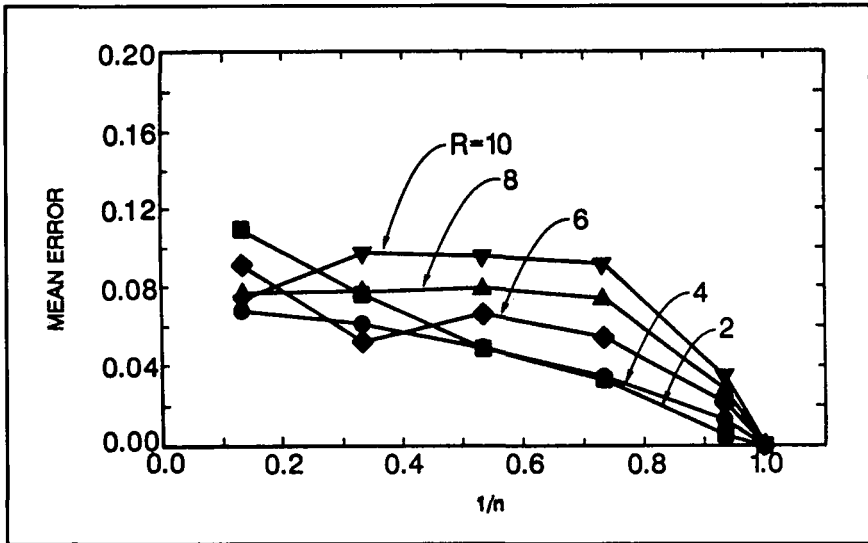Figure 6. Mean error versus 1/n at solute concentration
of 100 mg/l.

Figure 7. Mean error versus 1/n at solute concentration
of 450 mg/l.

was less than 13 percent over the linear retardance range modeled and decreased as the boundary concentration increased, while the greatest mean error resulted from the uncertainty in choosing the Freundlich exponent, 1/n. A mean error as high as 18.5 percent was identified at a boundary concentration less than 1 mg/l, but rapidly decreased to the 10 percent range as the boundary concentration increased.

The commonly applied adsorption approximation used in many groundwater contaminant transport codes relies on linear retardance. In general, where low values of contaminant are present, this is an acceptable approximation. Linear retardance, however, is a spatial and temporal constant and is readily estimated from Equation (10) given a few aquifer parameters. It is most notably independent of C, solute concentration. Nonlinear retardance, however, is dependent upon C which varies spatially and temporally along flow paths within an aquifer. Therefore, under conditions where non-linearity occurs, such as with high boundary concentration levels, errors can result from the selection of codes based on traditional retardance concepts.

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