

MODELING SOLUTE TRANSFER FROM SOIL TO SURFACE RUNOFF: THE CONCEPT OF EFFECTIVE DEPTH OF TRANSFER

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ABSTRACT

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A model for predicting the transfer of chemicals from soil to surface runoff water is described. The model applies a perfectly mixed reactor analog to an equivalent and homogeneous soil layer of uniform concentration near the soil surface, called the "Effective Depth of Transfer" (EDT). Solute concentrations in the EDT model are taken to be equal to the soil surface concentration as determined by an analytical solution of the one-dimensional diffusion equation for the limiting case of no infiltration. The model considers linear equilibrium sorption-exchange, and also accounts for rate-limited mass transfer through a laminar boundary layer at the soil surface-runoff water interface. The time-dependent EDT was found to vary as a linear function of the square root of time. The accuracy of the EDT approach is illustrated with one example, which compares the EDT solution for the soil surface concentration with the exact analytical solution of the diffusive mass transfer equation. The ability to change values of the physically based parameters adds considerable flexibility in application of the model to different locations with site-specific runoff regimes, soil properties, and solute characteristics.

INTRODUCTION

Pesticides, nutrients, and other agricultural chemicals transferred from soil to surface runoff water are potentially important pollutants of nearby surface waters. Predicting the release of soil-applied chemicals to surface runoff requires a model quantifying the interaction between the soil solution in the upper part of the soil profile and runoff water flowing over the soil surface. These interactions have generally been modeled by using a perfectly mixed reactor analog in which a chemical in the runoff water equilibrates instantaneously with that in the soil solution [e.g., see Ahuja (1986) for a review]. The approach assumes that the chemical is at all times uniformly distributed over a limited soil depth, which was called the "Effective Depth of Interaction" (EDI) by Ahuja et al. (1981). Most existing models assume that the EDI is independent of specific rainfall-runoff parameters, soil properties, and the type of chemical involved.

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Recorded data for rainfall-induced runoff show that runoff concentrations are initially high and later decline exponentially with time. This ideal behavior has been especially observed when simulated rainfall was applied to soils under controlled field conditions, or to soil boxes with permeable and impermeable bottoms (Ahuja and Lehman, 1983; Snyder and Woolhiser, 1985). Various attempts have been made to fit unknown parameters in exponential-type chemical runoff models to measured data. The effective soil mixing depth, or EDI, in particular has often been used as a fitting parameter. For example, Crawford and Donigian (1973), in their early model, assumed an active mixing depth of 0.3 cm at the soil surface. Donigian et al. (1977) later estimated this depth to be in the range of 0.2–0.6 cm by calibrating their model with experimental data. In many other models the thickness of the mixing zone was arbitrarily chosen to be 1 cm. Leonard et al. (1979) examined pesticide data from a number of watersheds and found runoff concentrations over a wide range of storm conditions to be strongly correlated with pesticide concentrations in the surface 1 cm of soil. Spencer et al. (1985) similarly examined pesticide concentrations in surface irrigation runoff waters from several large fields in the Imperial Valley of California. Their data also indicated that concentrations in the upper 1 cm of soil are good indicators for pesticide concentrations in furrow irrigation runoff water. The correlation for individual pesticides was found to be better than the correlation for combined classes of herbicides or insecticides.

Ingram and Woolhiser (1980), Ahuja and Lehman (1983), and Snyder and Woolhiser (1985) showed that solute concentrations in runoff water are usually much lower than those in the soil solution, even if measured immediately below the soil surface. Thus, complete mixing between surface runoff and soil water may not occur in practice. The observations by Snyder and Woolhiser (1985) seem to indicate that incomplete mixing is especially prevalent during low infiltration rates. To account for incomplete mixing between solution and surface runoff water, several investigators introduced additional degrees of freedom in their models. For example, Ingram and Woolhiser (1980) derived and evaluated a chemical extraction model which assumes incomplete mixing between soil and runoff water, but complete mixing for water infiltrating through the mixing zone. Frere et al. (1980) similarly used a partitioning coefficient between solute concentrations in runoff water and soil water in the upper 1 cm of soil.

Ahuja (1982) measured bromide concentrations in runoff during simulated rainfall on soil boxes with permeable and impermeable bottoms. The bromide concentrations in runoff from the saturated soil boxes with impermeable bottoms were several times higher than those from the permeable boxes. Ahuja was able to predict the concentration–time runoff hydrographs for the impermeable-box experiment using a well-mixed reactor model with mixing depths in the order of 50–100 cm or larger. As those depths were much greater than the actual soil depths used in his study, Ahuja questioned the concept of an effective thin mixing zone at the soil surface, at least for his experiments with the impermeable boxes. In subsequent experiments, also involving

simulated rain on soil boxes with impermeable bottoms, Ahuja and Lehman (1983) found soil bromide concentrations to increase very rapidly with depth, instead of being uniformly distributed as assumed in the mixing model. The bromide was lost to runoff water from depths of 2 cm and more. Ahuja and Lehman (1983) then suggested that the degree of mixing between rainwater and soil should be made an exponentially decreasing function of soil depth. The resulting empirical model closely matched the measured data for the impervious bottom boxes, but not for the pervious ones. These and other experiments suggest that the use of a fixed mixing zone near the soil surface will not lead to an accurate prediction of solute concentrations in both runoff and soil water.

Wallach et al. (1988) suggested that the release of dissolved chemicals from soil solution to surface runoff water should be viewed as a mass transfer process governed by both diffusion and convective transport with runoff water away from the soil surface. The transfer from soil to surface runoff water is essentially the same with rainfall as with lateral surface irrigation, although raindrop kinetic energy could enhance the transfer rate somewhat during rainfall. When infiltration is avoided by using soil boxes with impervious bottoms, the transfer rate should become dependent mainly on the runoff flow regime and the diffusivity of the dissolved chemicals. Thus, Wallach et al. (1988) could show that solute concentrations in runoff water, and in the soil profile itself, are described immediately by solutions of the diffusion equation subject to appropriate flux-type boundary conditions at the soil surface. Their model accurately predicted the runoff and soil solution concentrations measured by Ahuja and Lehman (1983) for the impervious boxes.

To understand more fully the interaction between soil and runoff water we shall in this paper also concentrate on the impermeable case without infiltration. Although lumping infiltration with other mass transfer processes helps in obtaining reasonable predictions for some runoff cases, it has so far not allowed models to be extrapolated to different locations by adjusting only a selected few model parameters. The combined diffusion-mass transfer solution of Wallach et al. (1988) will be used here to develop a new approach to the concept of an effective depth of soil which contributes to chemical runoff. The concept of an effective depth remains useful as it allows the inclusion of a relatively simple mass balance equation in existing models of the runoff process. The model could also serve as a simplified basis for more comprehensive, conceptually-based watershed models in the future.

THEORY

As mentioned above, experimental evidence suggests that chemicals in the soil solution near the surface are not uniformly distributed with depth during runoff. Therefore, a simple mass balance equation for an arbitrarily fixed soil surface layer cannot be written, and the advantage of having a well-mixed reactor with a relatively simple solution appears to be lost. However, we will show that, by making the effective upper soil layer time dependent, an

improved description results without losing the simplicity of a well-mixed reactor model.

Effective depth of transfer

We define an "Effective Depth of Transfer", EDT, as a surface soil layer in which, at each instance in time, the dissolved concentration is constant with depth and equal to the soil surface concentration as determined with the physically based model of Wallach et al. (1988). We assume that, in the absence of infiltration, chemicals are transported in the soil solution by diffusion, i.e.:

$$R \frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial z^2} \quad (1)$$

where c (mol m^{-3}) is the soil solution concentration, z (m) is the vertical coordinate (positive downwards), D ($\text{m}^2 \text{s}^{-1}$) is the porous medium ionic or molecular diffusion coefficient, and R (—) is a retardation factor accounting for linear equilibrium adsorption–desorption or exchange with the solid phase of the soil:

$$R = 1 + \rho K_d / \theta \quad (2)$$

in which ρ (kg m^{-3}) is the soil bulk density, θ (—) is the volumetric soil water content, and K_d ($\text{m}^3 \text{kg}^{-1}$) is the distribution coefficient between the soil matrix and the soil solution. The diffusion coefficient D in eqn. (1) accounts for the increased path length within the porous media, compared with diffusion in pure water. The Millington–Quirk tortuosity model was used successfully in the past (Jury et al., 1983; Wallach et al., 1988), and will also be used here:

$$D = \theta^{7/3} D_0 / n^2 \quad (3)$$

where D_0 ($\text{m}^2 \text{s}^{-1}$) is the ionic or molecular diffusion coefficient in water, and n (—) is the soil porosity.

A soil surface boundary condition at the soil–runoff interface must be formulated to describe the transfer of chemical from the soil to runoff water. Following Wallach et al. (1988), we describe this transfer by means of a rate equation characterizing the diffusion-controlled transport through a thin laminar boundary layer at the soil surface. The equation assumes that the solute concentration in runoff water is small compared with the soil solution concentration so that it has a negligible effect on the release rate from soil to runoff water. The approach leads to the following expression for the solute flux density, J_0 ($\text{mol m}^{-2} \text{s}^{-1}$), at the soil surface:

$$J_0(t) \equiv \theta D \frac{\partial c(0, t)}{\partial z} = \theta k c(0, t) \quad (4a)$$

where k (m s^{-1}) is a mass transfer coefficient for the laminar boundary layer. For convenience, J_0 is taken to be positive when directed upward from soil to surface runoff water. The magnitude of the mass transfer coefficient is controlled mainly by the diffusion coefficient of the chemical, and by such flow

characteristics as soil surface lateral slope, surface roughness, runoff water depth, and rainfall intensity and duration. It should be noted that k in eqn. (4a) is assumed to be finite. The limiting case when $k \rightarrow \infty$ characterizes instantaneous equilibration between soil and surface runoff water (no film resistance). Again neglecting the effect of a small but positive runoff concentration, the soil surface boundary condition is then reduced to:

$$c(0, t) = 0 \quad (4b)$$

The initial concentration, C_0 , in the semi-infinite soil profile is assumed to be constant, leading to the following initial and boundary conditions:

$$c(z, 0) = C_0 \quad (5)$$

$$\frac{\partial c}{\partial z}(\infty, t) = 0 \quad (6)$$

The solution of eqn. (1) subject to eqns. (4a), (5) and (6) is (Crank, 1967):

$$c(z, t) = C_0 \left\{ 1 + \exp\left(hz + \frac{h^2 Dt}{R}\right) \operatorname{erfc} \left[\frac{Rz + 2hDt}{2(DRt)^{1/2}} \right] - \operatorname{erfc} \left[\frac{Rz}{2(DRt)^{1/2}} \right] \right\} \quad (7a)$$

where $h = k/D$ (m^{-1}) and erfc is the complementary error function. When $k \rightarrow \infty$ (and hence $h \rightarrow \infty$), eqn. (7a) reduces to:

$$c(z, t) = C_0 \operatorname{erf} \left[\frac{Rz}{2(DRt)^{1/2}} \right] \quad (k \rightarrow \infty) \quad (7b)$$

where $\operatorname{erf} = 1 - \operatorname{erfc}$. Equations (7a) and (7b) predict soil solution concentrations which increase with soil depth in a manner consistent with the experimental results of Ahuja and Lehman (1983) and Snyder and Woolhiser (1985).

The solution concentration at the soil surface, c_s , follows from eqns. (7a) and (7b) by setting $z = 0$:

$$c_s(t) = C_0 \exp(\gamma^2) \operatorname{erfc}(\gamma) \quad (k < \infty) \quad (8a)$$

$$c_s(t) = 0 \quad (k \rightarrow \infty) \quad (8b)$$

where $\gamma = h(Dt/R)^{1/2}$. We note that the limiting case of an infinite mass transfer coefficient reduces to zero concentration at the soil surface, which is consistent with the imposed boundary condition [eqn. 4(b)]. Using eqns. (4a) and (8), we may write for the solute flux density from soil to runoff water:

$$J_0(t) = C_0 \theta k \exp(\gamma^2) \operatorname{erfc}(\gamma) \quad (k < \infty)$$

The solute flux across the soil surface for infinite k can be calculated from the concentration gradient at $z = 0$ as expressed by the first part of eqn. (4a). Substituting eqn. (7b) into the second term of eqn. (4a) and letting $z \rightarrow 0$ yields:

$$J_0(t) = C_0 \theta \left(\frac{DR}{\pi t} \right)^{1/2} \quad (k \rightarrow \infty) \quad (9b)$$

Equation (9b) follows also immediately from eqn. (9a) by letting h (and hence

k) go to infinity. Integration of eqns. (9a) and (9b) gives the total amount of solute, M (mol s⁻¹) released by the soil per unit cross-sectional area:

$$M(t) = \frac{C_0 \theta R}{h} \left[\exp(\gamma^2) \operatorname{erfc}(\gamma) - 1 + \frac{2\gamma}{(\pi)^{1/2}} \right] \quad (k < \infty) \tag{10a}$$

$$M(t) = 2C_0 \theta \left(\frac{DRt}{\pi} \right)^{1/2} \quad (k \rightarrow \infty) \tag{10b}$$

A mass balance for solutes in the EDT gives:

$$M(t) = \theta R H(t) [C_0 - c_s(t)] \tag{11}$$

where $H(t)$ (m) represents the effective depth of transfer. [It should be noted that $c_s(t)$ at all times is zero for the limiting case when $k \rightarrow \infty$.] Equation (11) shows that the EDT is equal to the cumulative amount of solute transferred from soil to runoff water at any time t , divided by the difference between the initial and the soil surface concentration at time t (the equation also includes the retardation factor to account for sorption or exchange when $R \neq 1$). Hence, the concentration profile at any point in time is approximated with a step function as shown in Fig. 1.

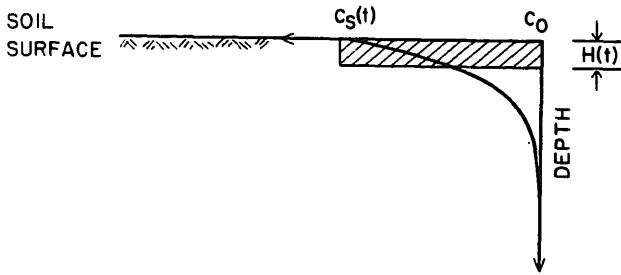


Fig. 1. Scheme of the equivalent depth of transfer (EDT) $\equiv H(t)$.

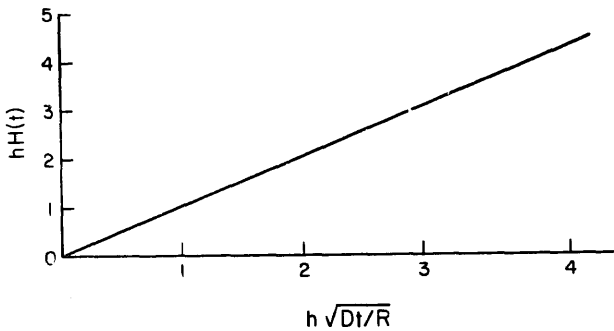


Fig. 2. Plot of hH versus dimensionless time, based on eqn. (12).

Substituting eqns. (8) and (10) into eqn. (11) and rearranging gives:

$$H(t) = \frac{1}{h} \left\{ \frac{2\gamma}{(\pi)^{1/2} [1 - \exp(\gamma^2) \operatorname{erfc}(\gamma)]} - 1 \right\} \quad (k < \infty) \quad (12a)$$

$$H(t) = 2(Dt/\pi R)^{1/2} \quad (k \rightarrow \infty) \quad (12b)$$

Figure 2 shows that $hH(t)$ is very close to being a linear function of γ . By using linear regression, with $r^2 = 0.9996$, the EDT as a function of the square root of time could be expressed as:

$$H(t) = -\frac{a}{h} + b(Dt/R)^{1/2} \quad (k < \infty) \quad (13)$$

with dimensionless coefficients $a = 0.0735$ and $b = 1.0725$. Because the variables in Fig. 2 are all dimensionless, the above linear relationship between the EDT and γ should be independent of the units chosen, and the soil properties or types of chemical involved. The value of b in eqn. (13) is about 5% less than the coefficient $2/(\pi)^{1/2} = 1.128$ in eqn. (12b) for the limiting case of no mass transfer resistance across the soil surface–runoff water interface. We note, however, that the EDT concept is based on the premise of a finite mass transfer process, and hence cannot be formally used for the limiting case given by eqn. (12b).

The dependency of the EDT concentration on time for different values of D , but for constant h and $R = 1$, is shown in Fig. 3. A constant $h = k/D$ means that all soil surface runoff features remain unchanged. The film model of Wallach et al. (1988) for determining the mass transfer coefficient shows that h^{-1} is equal to the thickness of the laminar boundary layer at the soil during runoff. By keeping h and other parameters constant, the expansion of the EDT in time can be examined with variations in each parameter.

We emphasize that the above EDT is fundamentally different from the EDI, used by Ahuja and Lehman (1983). Conceptually, EDI is based on the instantaneous mixing of raindrops with the soil solution in the upper part of the

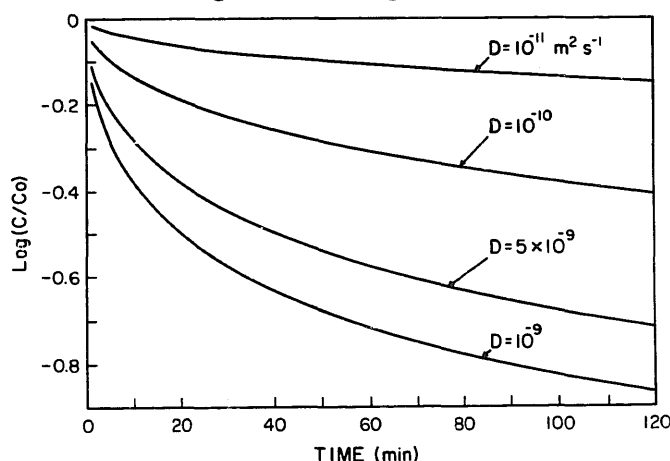


Fig. 3. Time dependency of the soil solution concentration in the EDT for various values of the diffusion coefficient: D ($R = 1$, $h = 0.15 \text{ m}^{-1}$).

profile. In contrast, EDT is based on more general considerations involving time-dependent diffusion and mass transfer through a water film at the soil surface. As such, EDT should be immediately applicable to different cases of rainfall- and irrigation-induced runoff, and to different locations in a watershed.

The EDT mixing model

The attribute of having a time-dependent but uniform concentration with depth enables the EDT to be easily incorporated into perfectly mixed, lumped-parameter models of the type described in the Introduction. To do this, concentration changes in the surface runoff water at the field outlet at different times may be calculated by representing the soil-runoff system by two interconnected well-mixed reactors. The upper reactor represents surface runoff water and the lower one the EDT. We again assume that runoff water concentrations are small compared with soil solution concentrations, thus invoking boundary condition (4b) at the soil surface. We also assume that water entering the system by rainfall or lateral irrigation is free of solutes. Hence, dissolved chemicals entering the runoff reactor are exclusively those released by the soil.

The mass balance equation for the EDT reactor is then:

$$R \frac{d}{dt} \{[c_s(t) - C_0] H(t)\} = -k c_s(t) \quad (14)$$

where, as before, k is the mass transfer coefficient (assumed from this point to be finite), $H(t)$ denotes the effective depth of transfer, and C_0 is the initial concentration. Equation (14) may be rearranged to give:

$$R \frac{d}{dt} [c_s(t) H(t)] = C_0 R \frac{dH(t)}{dt} - k c_s(t) \quad (15)$$

The first term of eqn. (15) expresses the change in the total amount of chemical stored in the EDT, and the second and third terms account for the transient expansion of the EDT to deeper soil depths and the release of solute from soil to runoff water, respectively.

No analytical solution for eqn. (14) is available when the exact relation eqn. (12) is used for $H(t)$. Instead, we will use the very accurate approximation given by eqn. (13). This equation predicts that the EDT will be negative during a very small time interval $0 \leq t < t_1$, where:

$$t_1 = \frac{a^2 DR}{b^2 k^2} \quad (16)$$

To avoid negative EDT values, we assume that the mass balance equation [eqn. (15)] is valid only for $H > 0$ and hence $t > t_1$. This assumption is most easily accounted for by introducing a new time coordinate $t' = t - t_1$ for t in eqn. (15), and approximating eqn. (13) with:

$$H = b \left(\frac{Dt'}{R} \right)^{1/2} \quad (17)$$

To maintain mass balance in the system we must then also introduce a new initial condition C'_0 for C_0 , which is obtained by evaluating eqn. (8a) at time t_1 . We note that t_1 depends on specific soil-chemical-runoff characteristics [see eqn. (16)], and hence must be calculated for each case separately.

Substitution of eqn. (17) into eqn. (15) leads to a first-order ordinary differential equation whose general solution is of the form:

$$c_s(t') = \frac{C'_0}{2\alpha} \frac{1}{(t')^{1/2}} + \frac{A}{(t')^{1/2}} \exp[-2\alpha(t')^{1/2}] \quad (18)$$

where $\alpha = h(D^{1/2}/b(\bar{r}))^{1/2}$ ($s^{-1/2}$) and A ($\text{mol m}^{-3} \text{s}^{1/2}$) is a constant which must be determined from the initial condition. Because of the singularity at $t' = 0$, A cannot be evaluated directly by substituting the initial condition into eqn. (18). Therefore, the initial condition for eqn. (15) will be taken as:

$$c_s(t'_0) = F C'_0 \quad (19)$$

where $t'_0 > 0$ is some small positive value of t' when the concentration in the EDT reactor can be determined, and $F < 1$ is the resulting fraction of the initial concentration C'_0 . F can be calculated immediately by noting that the uniformly distributed soil solution concentration in the EDT is also equal to the soil surface concentration as given exactly by eqn. (8a). For any arbitrary (small) value of t'_0 , F is therefore the ratio of $c_s(t'_0)$ as calculated with eqn. (8a) and C'_0 . This leads to the following expression for the soil solution concentration in the EDT:

$$c_s(t') = \frac{C'_0}{(t')^{1/2}} \left[\frac{1}{2\alpha} + \left(F(t'_0)^{1/2} - \frac{1}{2\alpha} \right) \right] \exp\{2\alpha[(t'_0)^{1/2} - (t')^{1/2}]\} \quad (20)$$

Figure 4 compares approximate results obtained with the above mixing cell

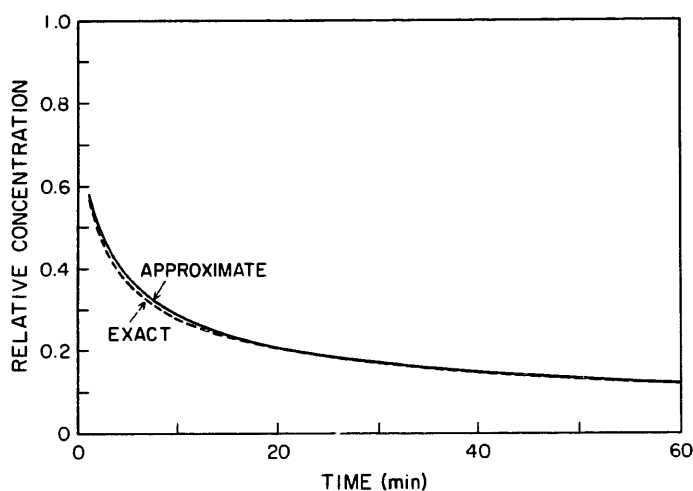


Fig. 4. Comparison of the exact [eqn. (8a)] and approximate [eqn. (20)] solutions for the solute concentration at the soil surface during runoff, using data listed in Table 1.

TABLE 1

Data for the Ruston fine sandy loam runoff experiment of Ahuja and Lehman (1983)

Parameter	Symbol	Value	Units
Rainfall intensity	P	1.89×10^{-5}	m s^{-1}
Slope length	L	1.0	m
Slope	S	0.04	—
Runoff water height	d	7.0×10^{-4}	m
Volumetric water content	θ	0.53	—
Initial concentration	C_0	4000.	g m^{-3}
Distribution coefficient	K_d	0.	$\text{m}^3 \text{kg}^{-1}$
Diffusion coefficient ^a	D	9.71×10^{-10}	$\text{m}^2 \text{s}^{-1}$
Mass transfer coefficient ^b	k	2.31×10^{-6}	m s^{-1}

^a $D = D_s/\theta$ as calculated by Wallach et al. (1988).

^b $k = k_l/\theta$ as calculated by Wallach et al. (1988).

approach with the exact results obtained with eqn. (8a) using the following data stemming from the experiments of Ahuja and Lehman (1983): $R = 1$, $D = 9.71 \times 10^{-10} \text{m}^2 \text{s}^{-1}$ and $k = 2.31 \times 10^{-6} \text{m s}^{-1}$ (Table 1). The plot shows that the approximate solution compares very well with the exact equation, thus indicating that eqn. (20) can be used with confidence for calculating EDT-concentrations. Equation (8a) was previously used by Wallach et al. (1988) in a well-mixed reactor model for the soil surface runoff component. That model successfully predicted the surface runoff concentrations measured by Ahuja and Lehman (1983) for soil boxes with impermeable bottoms. We conclude that the simplicity of eqn. (20), compared with eqn. (8a), makes the equation especially attractive for inclusion in more comprehensive chemical hydrograph models which also consider the residence time of the solute during runoff over a field.

SUMMARY AND CONCLUSIONS

The enhanced diffusion model of Wallach et al. (1988) was used to develop a new model for the transfer of chemicals from soil to runoff water. The model is based on the application of a perfectly mixed reactor analog to an effective soil layer near the soil surface, called the "Effective Depth of Transfer" (EDT). Solute concentrations in the EDT are taken to be equal to the solution concentration at the soil surface, c_s , as determined by an analytical solution of the one-dimensional diffusion equation subject to mass transfer through a laminar boundary layer at the soil surface-runoff interface. The time-dependent EDT was found to vary as a linear function of the square root of time. The EDT model can be combined with most lumped-parameter models used previously for predicting field effluent concentrations.

The model developed here should predict chemical runoff data better than is possible with most empirical models used thus far. Results obtained with the

EDT model compare well with the analytical solution of the solute diffusion equation. The ability to change the physically based parameters gives the model added flexibility in its application to different locations with case-specific runoff regimes, soil properties, and solute characteristics. The model does not at present include the infiltration of water into the soil, a phenomenon that is known to significantly affect the chemical runoff process. The infiltration case will be treated in another paper (Wallach and Van Genuchten, in prep.).

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