The Concept of Convective Mass Transfer for Prediction of Surface-Runoff Pollution by Soil Surface Applied Chemicals

Rony Wallach, William A. Jury, William F. Spencer

ABSTRACT

A model describing chemical transfer from soil to surface runoff is presented as a convective mass transfer process through a boundary layer connecting a soil mixing cell to the runoff mixing cell. The boundary layer is assumed to be laminar above the soil surface, restricting chemical transport through this zone to molecular diffusion. The determined mass transfer coefficient is proportional to the chemical diffusion coefficient and inversely proportional to the laminar boundary layer depth. It is increasing with the increase of soil surface roughness, runoff hydraulic gradient and runoff hydraulic radius.

A model for chemical runoff from concentration of pollutants in rainfall-induced surface runoff is represented as a well-mixed reactor. Transfer of solutes to the reactor is assumed to occur by a rate limited process proportional to the soil concentration. This approach differs from others in the literature which assume instantaneous equilibrium between runoff and soil solutions. Predicted outflow concentrations were calculated using both the equilibrium and the rate-limited models and the differences were analyzed. The main difference between the two models occurs during the early stage of runoff when the equilibrium model predicts lower chemical load of the outflow. The mass transfer model is characterized by two time scales that control the cumulative mass loss to the field outlet. One represents the runoff volume residence time and the other the diffusive soil mixing time.

INTRODUCTION

Modeling a system involving the adsorption and subsequent transport of chemicals from the soil to surface runoff water is a complex task. Many dynamic processes are involved, each one of which has a characteristic time scale over which it operates. Bailey et al. (1974) described the chemical transport process as consisting of four different mechanisms: (1) Diffusion and turbulent transport of dissolved chemical from soil by movement of soil water into overland flow, (2) Chemical desorption from solid phase adsorption sites into the soil water or directly into overland flow, (3) Dissolution of chemical from solid phases into the soil water or overland flow, and (4) Detachment and bulk transport of chemical present in the solid phase or chemical attached to soil particles or organic matter by hydraulic forces, followed by subsequent release of the chemical to the overland flow by dissolution or adsorption-desorption reactions. They also noted that the relative importance of each of these transport mechanisms is determined by the chemical under consideration, the method of application, soil characteristics, vegetation and recent hydraulic history. But generally, the rate of chemical transfer from soil into runoff is the critical step in modeling chemical runoff losses.

One way to analyze such a complex dynamic system is to formulate a series of coupled partial differential equations describing the different processes. For the system described above, a complete local dynamic description would involve a considerable computational burden and would require the solution of coupled nonlinear equations. Furthermore, such a description would necessarily contain large numbers of parameters whose values are not known a priori. Because of the complexity of the soil-runoff system, previous researchers have developed models only after making considerable simplifications in the description of the transport and exchange process.

Most investigators in the past have used a lumped parameter approach to describe the chemical extraction and transport process. In the simplified models which represent the soil-runoff water system under rainfall-induced runoff as a unit, the interaction between the soil solution phase and the runoff water is represented by an idealized mass exchange process which usually contains the following characteristics:

- There exists a fixed, thin mixing zone below the soil surface where rainfall mixes completely (Donigian et al., 1977; Steenhuis and Walter, 1980) or partially (Ingam and Woolhiser, 1980; Freer et al., 1980; Alujia and Lehmam, 1983) with soil solution.
- Mixing between rain water and soil water within the mixing zone is assumed to be instantaneous.
- Dissolved chemical in the system is partitioned between infiltration, runoff and soil water in proportion to the amount of water in each region.

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Contributors of Department of Soil and Environmental Sciences, University of California, Riverside, and U.S. Department of Agriculture, Riverside, CA.

The authors are: BODI WALLACH, Lecturer, Irrigation and Soil Sciences, Theshakeen Center for Soil and Water Sciences, The Hebrew University of Jerusalem, Jerusalem, Israel; WILLIAM A. JURY, Professor, Soil Physics, Department of Soil and Environmental Sciences, University of California, Riverside, CA; and WILLIAM F. SPENCER, Research Chemist, USDA-ARS, University of California, Riverside, CA.

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In these models the mass balance equation for the entire field has been formulated by regarding the soil solution and runoff water as a complex mixed reactor as given in equation [1]:

\[
\frac{d}{dt} [B_D + B_S] = -P
\]

where \( C \) is the chemical concentration in the soil solution and the runoff water, \( P \) is the constant rainfall rate, \( B \) is the height of runoff water above the soil surface, \( D \) is the thickness of the soil mixing zone and \( S \) is the volumetric water content of the soil.

The solution of equation [1] when the initial concentration is \( C_0 \) is:

\[
C(t) = C_0 e^{-kt} - P(B_0 - B_S)
\]

If infiltration is included in this calculation, an identical result is obtained when the completely mixed reactor model is used. Hence, any observed dependence of the runoff concentration distribution on infiltration rate should be taken as an indication that the assumption of complete mixing is not valid. Ahuja and Lehman (1983) studied rainfall-induced runoff of chemicals in soil tank experiments on which the infiltration rates were varied. They observed different concentration vs. time curves for different infiltration rates, and were not able to obtain good agreement with the completely mixed reactor model discussed above. To improve the mixing model, Ahuja and Lehman (1983) adopted the model used by Ingram and Wollheim (1980) in which incomplete mixing is assumed to occur both between the soil solution and runoff water and between the soil solution and infiltration water. For this model, the mass balance equation is written:

\[
\frac{d}{dt} \left[ B_D + B_S + \frac{1}{\kappa} \right] = \left( \frac{P}{\kappa} - C_0 \frac{1}{\kappa} \right) - \frac{d}{dt} \left[ \frac{1}{\kappa} \right]
\]

where \( \kappa \) is the ratio between the runoff water concentration and the soil solution concentration within the mixing zone, \( \kappa = \frac{1}{\kappa} \) is the ratio between the infiltration water concentration and the soil solution concentration within the mixing zone, \( \frac{1}{\kappa} \) is the runoff rain rate, equal to the difference between the rainfall rate and the infiltration rate. Ahuja and Lehman also used the approximation \( \kappa << \frac{1}{\kappa} \). The solution of equation [3] is given by equation [4]:

\[
C(t) = C_0 e^{-\frac{1}{\kappa} P(B_0 - B_S)}
\]

Although equation [4] predicted the experimental results of Ahuja and Lehman (1983) better than equation [2], the model still did not do a good job of describing the shapes of the runoff concentration curves. In order to achieve a better degree of agreement, these authors modified the mixing zone concept by assuming that the degree of mixing between the rainfall and the soil solution decreases with soil depth. This model produced a better agreement between the data and the prediction than any of the other models. However, this agreement was achieved by fitting the constant parameters to the data.
Thus, the mass transfer coefficient \( k_b \), which is by definition equal to the ratio of \( N \) to the mass flux between the two layers and the concentration difference, will depend on the following parameters:

\[
k_b = \frac{N}{DC} = f(D, C, \alpha, \beta, \gamma, \alpha_p, \beta_p, \rho_p) \tag{7}
\]

**Mass Transfer Coefficient**

The mass transfer coefficient from the soil to the runoff water will depend critically on the nature of the flow within the runoff phase, i.e., laminar or turbulent. If the depth of water over the surface is sufficient to produce persistent eddies, then the flow is classified as turbulent. However, if the depth of flow is smaller and the water velocity slower, then the viscosity of the fluid will control the flow and the flow will be laminar. Characterization of the mass transfer process becomes more complicated when rainfall is striking the surface of the runoff water (Lindley et al., 1982). In this case, the flow will be laminar close to the upstream boundary, but the portion of area covered by turbulent flow will increase downstream because of the increased depth and velocity of the runoff water. Thus, except over very uniform surfaces, the runoff flow is quite likely to be mixed between laminar and turbulent.

The fluid index which best characterizes the nature (laminar or turbulent) of the flow is the Reynolds number \( R_e \), which is a dimensionless number given by

\[
R_e = \frac{D u \gamma}{\mu} \tag{8}
\]

Any fluid flow with \( R_e \geq 500 \) is considered to be fully turbulent. This will be assumed to represent the runoff water.

**Transport of Mass from a stagnant region into a flowing turbulent region is best described using a boundary layer concept explained in detail in Kay and Noddorman (1955).** By adapting this model we assume that the water volume comprising the runoff over the surface can be divided into two regions: a laminar layer of thickness \( d \) adjacent to the soil surface and a turbulent, perfectly mixed region above it. This two-region model is also known as the film model. Within the stagnant boundary layer thickness \( d \), which is assumed to be much smaller than the average total runoff height \( H \), mass transfer is assumed to occur only by molecular diffusion. Thus, a drop in concentration occurs over this layer. Above the laminar layer, the turbulent region has a uniform concentration in the vertical direction.

In practice, the transition between the laminar and turbulent regions is not sharply defined and the film model only approximates conditions near the boundary (Kay and Noddorman, 1955).

As a result of the assumption the mass transfer through the boundary layer occurs by diffusion, we may write the mass flux \( N \) using Fick's law as

\[
N = \frac{D (C_{o} - C_{w})}{6} \tag{9}
\]

where \( D \) is the liquid diffusivity for the chemical in water, \( C_{o} \) is the chemical concentration at the soil surface and \( C_{w} \) is the uniform chemical concentration in the turbulent region, which may be regarded as the runoff water concentration \( C_{w} \).

Thus, the mass transfer coefficient for the film model, defined in equation (7), may be set equal to

\[
k_b = \frac{\rho \gamma}{DC} \tag{10}
\]

It will be of great interest to relate the mass transfer coefficient to the runoff flow parameters in order to obtain a measure of the expected chemical flux leaving the soil surface.

As mentioned above, in the film model, the flow system is considered as consisting of two separate layers: the main turbulent flow layer in which viscous stresses are insignificant in comparison to the Reynolds stresses, and the viscous layer in which the turbulent motion is damped out by the action of viscosity. In the viscous layer, the shear stress must ultimately be transmitted in the same manner as in the laminar boundary layer, i.e., as a product of the coefficient of viscosity and the mean velocity gradient using Newton's Law of Viscosity:

\[
\tau = \frac{\mu \omega}{6} \tag{11}
\]

If it is assumed that the shear stress \( \tau \) remains constant in both film layers, then the shear stress described in equation (11) is equal to the shear stress at the soil surface. If it is also assumed that the velocity varies linearly in the viscous film, equation (11) may be written as

\[
\tau = \frac{\mu \omega}{6} \tag{12}
\]

where \( \omega \) is the mean velocity above the laminar boundary layer.

By equating the frictional forces along the wettled wall of an elementary stretch of channel to the component of the gravitational force in the direction of the flow, the shear stress may be written as

\[
\tau = \frac{gD \rho}{6} \tag{13}
\]

where \( \rho \) is the density of water, \( g \) is the acceleration of gravity, \( R \) is the hydraulic radius (the ratio of the wetted area to the wetted perimeter) and \( J \) is the hydraulic gradient.

From the channel flow hydraulics it is known that shear stresses are related to the friction velocity \( u \) by (Bird, Stewart and Lightfoot, 1960):

\[
\frac{u}{\sqrt{gD}} = \frac{\psi}{\rho} = (gD)^{1/2} \tag{14}
\]

After eliminating \( d \) in equation (12) and (10) and multiplying through by the hydraulic radius \( R \), one obtains:

\[
\frac{\tau}{\mu \omega} = \frac{R}{\rho} \frac{\tau}{\mu \omega} \tag{15}
\]

The right side of equation (15) is the Sherwood number.
Rearranging the left side of equation [15] yields:
$$\frac{\rho_d^3}{\mu^2} = \frac{1}{2} C_d R_e$$
where
$$R_e = \frac{\rho_d R}{\mu}$$
is the Reynolds number and
$$C_d = \frac{2 \ln \frac{R}{d}}{u_2}$$
is the friction factor. Combining [14] and [18] produces
$$C_d = \frac{2 \ln \frac{R}{d}}{u_s}$$

The Sherwood number may also be written as using equation [10]:
$$S_h = \frac{k_d R}{D} = \frac{R}{D}$$

Hence, the viscous film thickness is $d$ may be written as:
$$d = \frac{2R}{C_{PF} S_h}$$

In order to define the film thickness in terms of parameters which are more easily measured, one substitutes equation [19] into equation [21], obtaining:
$$d = \frac{2R}{C_{PF} \sqrt{rac{2 \ln \frac{R}{d}}{u_s}}}$$

If one substitutes the explicitly Manning equation for the average cross-sectional velocity (Linsley et al., 1983):
$$u_m = f R^{1/2} R^{-1.25}$$
and the Reynolds number equation [17] for channel flow into equation [22], one obtains:
$$d = \frac{1}{n \cdot \sqrt{\frac{f L}{R}}} \frac{R}{D}$$

where $n$ is the Manning roughness coefficient. For pluvial flow it may be assumed that the hydraulic radius $R$ is equal to the water depth $d$ over the surface.

As seen from equation [24], the mass transfer coefficient will increase as the soil surface roughness $R$, the hydraulic gradient $J$, and the hydraulic radius $R$ or water depth increase. These conclusions are consistent with the laboratory findings of Ahuja (1982), and Parr and Lane (1985), in a series of experiments conducted in order to study the effects of slope, field length, rainfall intensity and soil surface roughness on soil the chemical transfer to runoff water.

As a numerical example, consider a long field with an average slope of 0.5% ($J = 0.005$) and an average water height during a storm of 0.1 m. For plume flow the Manning roughness coefficient is approximately $n = 0.02$. Using $\rho = 1.02 \times 10^3$ kg m$^{-3}$, $\mu = 1000$ kg m$^{-1}$ s and $g = 9.81$ m/s$^2$, equation [24] gives $d = 1.56 \times 10^{-4}$ m.

### MODEL APPLICATION

The rate-limited mass transfer concept will be now combined with two different models, to predict the chemical concentration distribution at the field outlet, in order to demonstrate the effect of the rate-induced transfer. In the first case, both models—the convective mass transfer and the one in which instantaneous equilibrium between soil solution and runoff water concentrations is assumed, are combined with the lumped soil surface layer model. The differences in the concentration distribution will be analyzed. As pointed out in equation [5], the mass transfer is only a link in a broad model for prediction of chemical concentration profiles at the field outlet. In the second case we shall deal with the expected concentration profiles when the rate limited mass transfer will be combined with two different cases of water application induced runoff (Wallisch et al., 1988).

#### Case 1: Mass transfer as a first order reaction.

Experiments had indicated that the chemical distribution of a certain soil surface layer is the main source of chemicals leaving the field with the runoff water during and after a given rainfall storm. This concept has been widely used in lumped soil surface layer models as was mentioned earlier. It is of an interest to contrast the expected concentration hydrographs at the field outlet when a certain time scale characterizes the transfer process as opposed to the instantaneous equilibrium.

If we assume that the chemical concentration within the soil solution greatly exceeds the concentration in the runoff water, and that the soil concentration $C_s$ is well mixed at all times up to a depth $d$, then equation [9] is reduced to:
$$\frac{dC_s}{dt} = -u_s C_s$$

which decouples the soil solution concentration from the runoff concentration. As a consequence, there is no spatial dependence of the soil concentration and the solute balanced equation for the entire soil zone, and mass balance for the soil surface layer is:
$$\frac{\partial C_s}{\partial t} = -u_s \frac{dC_s}{dt}$$

with $C_s(0) = C_s$. Equation [26] has the solution:
$$C_s(t) = C_s \exp \left( -\frac{t}{u_s} \right)$$

anywhere on the field.

Combining equation [26] with the mass balance equation for the soil-runoff system, equation [1] gives:
$$\frac{d}{dt} \left[ H C_s - \beta \left( C_s \right) \right] = -H C_s \frac{dC_s}{dt} - k_d C_s$$
where $C_w$ is the runoff water concentration. Using equation (27), equation [28] may be written as:

$$\frac{dC(t)}{dt} = -P_e C(t) + k t C(t) \exp(-k_t t/\theta D)$$  

$$\text{where } C(0) = 0.$$  

Equation [29] has the solution:

$$C(t) = \frac{h_0 C_0}{P_e + k t \theta D} \exp(-k_t t/\theta D) \exp(-Pt/t)$$

This solution is plotted in Fig. 1b as a function of dimensionless time $\tau = P_t/t$ for various values of $B = \text{KL}/P$, assuming $w = 60\text{H}/H = 10$ (a deep soil layer) and in Fig. 2b for $w = 2$ (a shallow soil layer).

There are two time scales influencing the movement of solute to the effluent. The longer time scale is associated with the rate-limited transfer of solute from the soil to the runoff, and is characterized by an effective mixing time $\tau_0 = 60\text{H}/K$. This time is reflected in the long effluent tail in Fig. 1b and 2b. The shorter time scale, characterized by $\tau_1 = H/P$, is associated with the residence time of the fluid in the runoff water since it leaves the soil. As the mass transfer time $\tau_0$ decreases relative to $\tau_1$, the curve develops a higher maximum (as represented by large values of $k$), corresponding to a rapid release of chemical from the soil to the runoff.

As mentioned above, several existing models of the soil-runoff system use the assumption:

$$C(t) = \text{OC} \quad (0 \leq \theta \leq 10)$$

This allows for the coupling of the soil and runoff water zones. Assuming that runoff is induced by a uniform rainfall rate $P$, the runoff is a well-mixed reactor cell, and that no infiltration occurs, the mass balance equation for the soil-runoff system is given by:

$$\frac{d}{dt}[H C(t) + \frac{\partial}{\partial t} \frac{dC}{dt}] = -P_e C(t) = (t + \frac{\partial}{\partial t}) \frac{dC}{dt}$$

where the quantity in brackets is the mass/volume of solute in the field (in both soil and runoff water) and $P_e$ is the rate of flow by lateral runoff (all per unit length of field).

Since it is assumed that a concentration $C_w$ is initially present in the soil solution prior to runoff, the initial runoff concentration is equal to:

$$C(t) = \text{OC} \quad H = 0$$

The solution to equation [32]-[37] is given by:

$$C(t) = \text{OC} \quad H = 0$$

Equation [34] is plotted in Fig. 1a as a function of dimensionless time $\tau = P_t/t$ for various values of $w_1$, assuming $w = 10$ and $w = 2$ in Fig. 2a.

The contrast between cases a and b in Fig. 1 and 2 is interesting. The area under each curve is identical, since both processes will ultimately result in a complete depletion of the chemicals from the surface layer of soil.

However, the equilibrium model, equation (26) (case b) predicts that the maximum concentration will occur at $t = 0$, whereas the maximum value for the rate-limited mass transfer model (case a) occurs some time later. The principal difference between the two models is the early time behavior. Therefore, if only the values of runoff effluent data are available, each model can be adjusted to fit to data with a comparable degree of precision.

There are significant management implications associated with the incorrect usage of equation [34] relative to equation [30], since the latter will overestimate the extent of leaching in the early stages because the soil and runoff volumes are complete mixed at $t = 0$. The only effect of modifying the proportionality constant $\theta$ is

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Case 2: Comparison of concentration hydrographs for lateral irrigation versus rainfall induced runoff.

An essential difference in the behavior of rainfall induced runoff flow as compared to the lateral irrigation flow is their residence time distribution (RTD) (Wallach et al., 1988a). The RTD, which is a probability density function, defines the fraction of flow that has residence times between $t$ and $t + dt$ approaches zero. The RTD for a steady rainfall induced runoff is equal to the RTD of a well-mixed reactor (Wallach et al., 1988a):

$$f(t) = \frac{1}{t} \exp(\frac{t}{r})$$

where $r = P/H$ is the average residence time, $P$ is the rainfall intensity and $H$ is the average water depth. On the contrary the RTD for a steady state surface irrigation induced runoff, in the absence of axial-convective mixing, is the delta function distribution defined as:

$$f(t) = \delta(t) = \int_{-\infty}^{\infty} \delta(t-\tau) \, d\tau = 1$$

where $\tau = L/V$ is the travel time required for a unit water volume entered at the upstream end to reach the effluent boundary, $L$ is the channel length, and $V$ is the uniform water velocity. The dissolved chemical flux, from the soil to the runoff water is (Parr et al., 1987; Wallach et al., 1988b):

$$j(t) = C_{i0}K_L \exp(\frac{t}{r}) \text{erfc}(\sqrt{t})$$

where $C_{i0}$ is the initial soil concentration, $K_L$ is the mass transfer coefficient, $r = k_i/DR$, $D$ is the soil liquid diffusion coefficient, and $R$ is the retardation factor as defined in Wallach et al. (1988b).

Wallach et al. (1988b) applied the convolution theorem between equation [35] and [37] to obtain the concentration hydrograph at the field outlet as:

$$(C_{out}(t)) = \frac{C_{i0}K_L}{P(1 + \gamma)} \left[ \exp(\gamma t) \text{erfc}(\sqrt{\gamma t}) - \sqrt{\gamma t} \exp(-t) \phi(\gamma t) - \exp(-t) \right]$$

where erf(...) is the imaginary error function and $P$ is the rainfall rate. Equation [38] can be easily evaluated by making use of a general subroutine (van Genuchten, 1985) for computing the complex error function.

Reapplying the convolution for lateral irrigation induced runoff, the concentration hydrograph at the downstream end is:

$$(C_{out}(t)) = \frac{1}{H} \int_{0}^{t} j(t') \phi \left( \frac{t}{H} \right) dt'$$

for $t < r$

$$(C_{out}(t)) = \frac{1}{H} \int_{t-r}^{t} j(t') \phi \left( \frac{t}{H} \right) dt'$$

for $t > r$

Introducing equation [27] into equation [39] yields

$$(C_{out}(t)) = \frac{C_{i0}k}{r} \left[ \exp(\frac{t}{r}) \text{erfc}(\sqrt{\gamma t}) - 1 - \frac{2\sqrt{\gamma t}}{\sqrt{\pi}} \right]$$

for $t < r$

$$(C_{out}(t)) = \frac{C_{i0}k}{r} \left[ \exp(\frac{t}{r}) \text{erfc}(\sqrt{\gamma t}) - \exp(-t) \phi(\sqrt{\gamma t}) + 2\sqrt{\gamma t} \phi(\sqrt{\gamma t}) - \sqrt{\gamma t} \exp(-t) \right]$$

where $H$ is the water depth.

Although the runoff water depth for surface irrigation-induced runoff must be larger than the rainfall-induced runoff in order to produce a given discharge at the end of the field, identical depths, $H$, were assumed for the illustration of similarities and differences between the two runoff cases.

The two concentration hydrographs are shown in Fig. 3. The shape of the concentration hydrographs seems to be made up by two main contributions. For a time interval smaller than $r$ (average residence time of runoff water) the shape is mainly controlled by the surface runoff RTD. The shapes for averaged times is controlled mostly by the rate of chemical transfer from soil surface.
to the runoff. Therefore, the concentration profiles for early times are different due to the differences of runoff RTD equations (35) and (36). Since the concentration profiles depend mainly on the mass transfer rate for \( \sigma \), the profiles are almost coincident for this time. The maximum concentration of the lateral water application induced runoff is higher as opposed to the decaying part where the concentration associated with the rainfall induced runoff is higher.

It should be mentioned that the concentration profiles in Fig. 3 were calculated for steady runoff flow. In reality, chemicals are also transferring during the transient runoff flow prior to the steady flow. Therefore, the shape of the concentration hydrographs which intend to simulate field or laboratory studies should include the contribution of early times of the initial runoff concentration. Their shape is of a decaying curve which initiates at a high concentration and coincides after a time lag of order \( \tau \) with the curves of Fig. 3, as shown in Wallach et al. (1988b).

SUMMARY AND CONCLUSIONS

The rate-limited chemical transfer concept which describes a diffusion based exchange of chemical between the soil solution and the overlying runoff water was presented. This concept was compared with the widely used equilibrium model, partitioning between a soil mixing zone and runoff water, for predicting outflow concentrations for rainfall-induced runoff. It was shown that, although the latter stages of the chemical hydrographs have similar behavior, the early time picture is quite different. Under rate-limited exchange, both the time scale of the rate-controlled release from soil to runoff and the time scale of the fluid residence time influence the cumulative release of chemical to the outlet end of the field.

The convective mass transfer model has also been used to compare between shapes of the expected concentration hydrographs at the runoff outlet due to rainfall- and lateral irrigation-induced runoff. The two runoff cases were found to differ mostly during the initial stages of the process, especially for time less than the average residence time of runoff water on the field. At these times, the runoff flow characteristics affect the concentration shape. On the contrary, for long times the overall rate is controlled by chemical transfer form the soil surface and the transport process has less effect on the concentration hydrograph shape. The maximum concentration, at the runoff outflow, is expected in the case of lateral irrigation induced runoff. The mass transfer coefficient, as based on the film model, was related to hydraulic characteristics. Its value will increase with soil surface roughness, \( \kappa \), the hydraulic gradient and the hydraulic radius increase. The model at present does not include the infiltration of water into the soil, a phenomena that is known to affect the chemical runoff process. The infiltration case will be treated in the future.

References