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CHEMICAL TRANSFER INTO OVERLAND FLOW

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ABSTRACT

The effects of rainfall intensity, soil surface slope, and rainfall kinetic energy upon the release of a chemical (CaSO_4) from the soil solution into surface runoff were examined by an analysis of variance and application of a mathematical model. The analysis of variance was performed upon an estimated depth of mixing at the soil surface. The mixing depth is assumed to be a zone in which the rain, surface flows, and chemical intermix. The mathematical model used was derived from mass balance considerations assuming exchange between the soil water in the mixing zone and overland flow. Results of the analysis of variance show significant interaction effects of the rainfall intensity, soil surface slope, and rainfall kinetic energy on the depth of the mixing zone. Analysis of the collected data in the framework provided by the mathematical model led to the observation that the chemical exchange rate between the mixing zone and overland flow is affected by these factors. The concentration of a chemical species in the overland flow was much lower than that of the pore water in the mixing zone.

INTRODUCTION

The 1972 Amendments of the Federal Water Pollution Control Act, which stressed control of pollution from nonpoint sources, stimulated the development of several agricultural chemical transport models (Crawford, et. al. 1973, Donigian and Crawford, 1976, Donigian, et. al. 1977, Bruce, et. al. 1975, Frere et. al. 1975). In each of these models, the component dealing with water and chemical transport across the soil-surface runoff interface is crucially important in understanding and predicting transport of weakly and moderately adsorbed chemicals. Surprisingly, a review of these papers revealed that the mathematical structure of this component in existing models is based upon very simplified assumptions. These assumptions are, perhaps, reasonable for a first approximation; however, no controlled experimental work has been done to observe the nature of chemical release into overland flow disturbed by raindrops.

The objective of this paper is to describe an experimental investigation of chemical transfer from a saturated soil into overland flow disturbed by raindrops and to examine the degree to which the experimental data agree with predictions of several alternative models of this important component of chemical transport models.

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DESCRIPTION OF THE CHEMICAL TRANSFER PROCESS

Bailey et al. (1974) described the chemical transfer processes involved in the pickup of pollutants (pesticides) by overland flow during a rainfall event. They identified the four mechanisms depicted in Figure 1: 1) liquid-liquid diffusion of the dissolved chemical by movement of soil water into the overland flow, 2) desorption of the chemical from soil particles into the soil water or directly into overland flow, 3) dissolution of solid phase chemical into the soil water or into overland flow, and 4) scouring of solid phase chemical by hydraulic forces and subsequent transport and moving dissolution. The first mechanism can be expanded to include the dispersion and turbulent transport of a solute into overland flow incurred by raindrop impact.

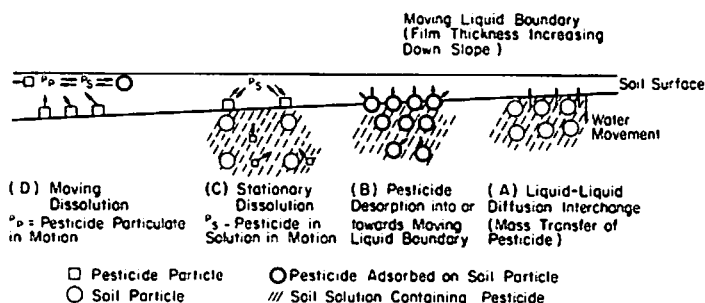


Figure 1. Modes of pesticides transport into and within the moving liquid boundary during a rainfall event. [From Bailey, Swank and Nicholson (1974)]

The relative importance of each of these mechanisms depends upon the particular chemical and the method of application and may change from one runoff event to another. However, a chemical present in the soil water solution as a result of desorption or dissolution from the solid phase can only move to the surface by mechanism 1 during a runoff event. Consequently, it appears that the diffusion and turbulent transport mechanism is significant. Movement of water and chemicals near the soil surface could be described mathematically with a one dimensional partial differential equation for convective dispersion with the dispersion coefficient increasing near the surface to account for the effects of raindrop impact. Past investigators have taken a simplified approach, however. Some have assumed that the exchange process is completely controlled by erosion so that the surface film of water and the suspended sediment with adsorbed chemicals is considered to be equivalent to a completely mixed reactor. (Huff and Kruger, 1967, Crawford and Donigan, 1973). More recently the concept of a "mixing zone" has been introduced by Steenhuis, et. al. (1978). Conceptually this zone is a depth of soil where rain, shallow flow, soil, soil water, and chemicals are mixed. The chemical within this zone may be in the solid or liquid phase, or both, or may be adsorbed on soil particles. This model attempts to take the dispersion process into account, but makes the rather unrealistic assump-

tion that the concentration of the chemical in the "mixing zone" is the same as the concentration in surface runoff.

An alternative formulation is developed in this paper.

Derivation of Model for Mass Exchange
Between a Mixing Layer and Surface Runoff

The following assumptions were made in deriving the model:

- i) Sediment transport is negligible and therefore the chemical transfer is only in the solution phase.
- ii) A finite mixing depth exists in which rain, shallow surface runoff, soil, soil water and chemicals are mixed.
- iii) Chemical transfer occurs by exchange of solution between the mixing zone and overland flow and by infiltration loss from the mixing zone.
- iv) The chemical concentrations in overland flow and in soil water of the mixing zone differ and each varies with time and distance.
- v) The concentration of the chemical in rainwater is negligible.
- vi) Overland flow is unsteady and spatially varied.
- vii) The infiltration rate is spatially uniform, but unsteady.
- viii) During a runoff event there is no movement of chemical from the soil below the mixing zone into the mixing zone.

A definition sketch for the problem is shown in Figure 2. The problem to be described is as follows: Surface runoff is occurring from a plane of length L_0 and slope S_0 , the rainfall rate is r and

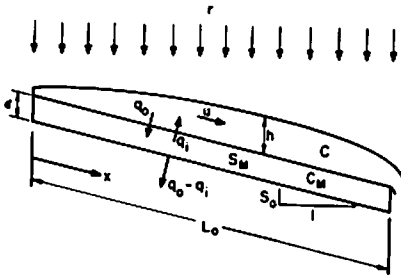


Figure 2. Definition Sketch

the local infiltration rate is $(q_0 - q_i)$ where q_0 is the rate of inflow of surface water into the soil and q_i is the rate of flow from the soil water into the surface runoff. The exchange rate, q_i is induced by raindrop impact and hydrodynamic forces. The local depth of surface runoff is h and the local vertically averaged velocity is u . The depth of the mixing zone is ϵ and the vertically averaged concentration of the chemical species is denoted by C in the surface runoff and C_m in the mixing zone. S_m is the source term representing the rate of dissolution of solid phase chemical and/or the desorption rate within the mixing zone.

By applying conservation of mass equations for water and chemical to differential elements of overland flow and mixing zone the following equations can be derived (Ingram, 1979).

$$h \frac{\partial C}{\partial t} + uh \frac{\partial C}{\partial x} + C \left(\frac{\partial uh}{\partial x} + \frac{\partial h}{\partial t} \right) = C_m q_i - C q_o \quad (1)$$

and

$$c \frac{dC_m}{dt} + C_m \frac{dc}{dt} = S_m + q_o (C - C_m) \quad (2)$$

Equation (1) expresses the mass balance in surface runoff without longitudinal dispersion and equation (2) expresses the mass balance in the mixing zone. No x-derivatives of C_m appear because it is assumed that no lateral flow occurs in the mixing zone. The expression in the parenthesis in Eq. (1) is the left hand side of the continuity equation for overland flow.

$$\frac{\partial h}{\partial t} + \frac{\partial uh}{\partial x} = r - (q_o - q_i) \quad (3)$$

If this expression is substituted into Eq. (1) we obtain

$$h \frac{\partial C}{\partial t} + uh \frac{\partial C}{\partial x} = q_i (C_m - C) - Cr \quad (4)$$

Equations (2) and (4) along with the appropriate initial and upstream boundary conditions constitute the model to be investigated.

Because these general equations are quite complicated for a preliminary investigation, we examined a simpler system by assuming the following:

- i) The equivalent mixing zone depth, c , is constant during a storm. Therefore $\frac{dc}{dt} = 0$.
- ii) The soil is initially saturated and the infiltration rate is zero. Therefore $q_o = q_i$.
- iii) There is no source for the chemical within the mixing zone, $S_m = 0$.

With these simplifications equations (2) and (4) become:

$$c \frac{dC_m}{dt} = q_o (C - C_m) \quad (5)$$

$$\text{and } h \frac{\partial C}{\partial t} + uh \frac{\partial C}{\partial x} = q_i (C_m - C) - Cr \quad (6)$$

EXPERIMENTAL FACILITY

The facility used for this study consisted of a sheet metal flume with a horizontally oscillating rainfall module mounted above (Ingram, 1979). The flume held a volume of soil with a surface area of 2 x 24 in (5.1 x 62 cm) and a depth of 2.75 in (7 cm). Slope adjustments were made by raising or lowering the flume supports; finer adjustments could be made by using a screw attached to the bottom of the supports. The rainfall module generated drops through capillary tubes 0.009 inch (.0229 cm) in diameter. The intensity of rainfall was varied by changing the height of a constant head tank relative to the module. To achieve a more uniform areal coverage, the module was oscillated in the horizontal plane by

a gear motor statically attached to the module support frame with its shaft attached to the free swinging rainfall module. This gear motor produced a circular movement with a radius of 0.25 in (0.635 cm) at 6 rpm.

In the derivation of the mathematical model, the presence of a chemical source within the mixing zone was accounted for; however, to simplify the model, the source term was set equal to zero. Therefore a relatively inert sand was used to prevent interaction with the saturating solution.

After the sand was placed in the flume (at a constant porosity of 41 percent in all experiments) the soil voids were saturated with a saturated CaSO_4 solution as a tracer. The CaSO_4 solution was siphoned from an elevated container to a 0.25-in (0.635-cm) perforated section of plastic tubing laid on the bottom of the flume. This saturated the soil from the bottom of the flume giving greater assurance of complete soil saturation.

Surface runoff was collected at the lower end of the flume and measured volumetrically in a collection stand pipe with a pressure sensing device that recorded the volume on a paper tape at 5-sec intervals.

EXPERIMENTAL INVESTIGATION

The first phase of this investigation consisted of testing the null hypothesis that the rainfall intensity, soil surface slope, and rainfall kinetic energy each has no effect upon the mass transfer of a chemical species into overland flow. The mathematical model derived suggested that this hypothesis could be tested by examining the effect of these variables on the equivalent depth of the mixing zone, ϵ .

The depth of the mixing zone, ϵ , for an experimental run could not be measured directly; therefore, a variable related to the mixing depth was used in this analysis. The estimate of this surrogate variable (ϵ_{30}) was based upon the assumptions that 1) the equivalent mixing depth (ϵ) varies neither with flow length nor time and 2) the concentration of CaSO_4 in the pore water after a 30-minute run is equal to the concentration in the surface runoff at that time. Although the second assumption is not consistent with the assumption used in the derivation of the mathematical model, the difference between C and C_m should become small as the duration of the storm increases; therefore, we assumed that C equals C_m after 30 minutes.

The storage of CaSO_4 within the mixing zone at any time can be written as:

$$M_m = C_m A \epsilon' \phi \quad (7)$$

where M_m = mass of CaSO_4 within the mixing zone (M)
 C_m = concentration of CaSO_4 in the mixing zone's pore water
 (ML⁻³)
 A = horizontal area of the mixing zone (L²)
 ϕ = saturated porosity of the soil
 ϵ' = effective depth of the mixing zone, $\epsilon' = \epsilon/\phi$. (L)

The change of mass within the mixing zone for a time interval is determined from the difference between the mass present at the start and that present at the end of the interval. This may be represented by the expression:

$$\text{Outflow of Mass} = M_{m1} - M_{m2} \quad (8)$$

where M_{m1} = mass within the mixing zone at the storm's beginning
 M_{m2} = mass within the mixing zone at the end of the storm, and by applying Eq (7), this becomes:

$$\text{Outflow of Mass} = (C_{m1} - C_{m2}) A \epsilon \phi \quad (9)$$

where C_{m1} = chemical concentration in the pore water of the mixing zone at the beginning of the time interval (ML^{-3})
 C_{m2} = chemical concentration in the pore water of the mixing zone at the end of the time interval (ML^{-3}).

The other terms are as previously described.

The tracer concentrations needed in Eq (9) were estimated from electrical conductivity (EC) measurements of the surface runoff. These electrical conductivity measurements were converted to concentrations of $CaSO_4$ by using a regression equation derived from EC measurements of salt solution samples:

$$\text{Salt Con. (g salt/100 g water)} = 0.0000351 EC^{1.13} \quad (10)$$

($r = 0.999$ and $S_{y,x} = 0.0129$)

The index value of the mixing depth (ϵ_{30}) was calculated from Eq. (9). This was possible because C_{m2} , C_{m1} , A and ϕ were recorded values and the total mass outflow could be estimated from the concentration vs. time curves and the hydrographs of each experimental run.

STATISTICAL ANALYSIS

The values of ϵ_{30} determined for each experimental run were used in an analysis of variance to test the null hypothesis. By this analysis the total variation of a sample may be partitioned into components according to possible sources of variation and the importance of each of these sources can be tested.

The objective of this analysis was to test the effect of rainfall intensity, soil surface slope, and kinetic energy on ϵ_{30} . The experimental runs were made with two rainfall intensities [2.2 inches/hr (5.6 cm/hr) and 3.6 inches/hr (9.1 cm/hr)], three soil surface slopes (0, 3, and 6 percent), and two kinetic energies controlled by two heights of fall [12 inches (31 cm) and 28 inches (70 cm)]. (In the rest of this paper, the notation of i_1 and i_2 ; S_1 , S_2 , and S_3 ; H_1 and H_2 shall be used for the values of the rainfall intensity, surface slope, and heights of fall with the higher subscripts representing the larger values). The values of ϵ_{30} for the experiments were calculated using the procedure described above and are presented in Table 1.

Using the information found in an analysis of variance, the null hypothesis--that the variance estimates are each independent estimates of the same quantity--was tested. An acceptance of the null hypothesis suggests that the experimental variables and interaction terms have no detectable effect on the mixing depth. To test the null hypothesis, an F test was used.

When performing the F test upon the variance estimates, the first test should be made upon the highest order interaction ($i \times S \times H$). If the variance estimate of this interaction proves to be significantly

WATERSHED MANAGEMENT

Table 1. Estimated values of ϵ_{30} (cms.) for each of the experimental runs. (The higher subscripted values denote the greater intensity (i), height of fall (H), and soil surface slope (S)).

	ϵ_{30}											
	i_1						i_2					
	H_1			H_2			H_1			H_2		
	S_1	S_2	S_3	S_1	S_2	S_3	S_1	S_2	S_3	S_1	S_2	S_3
	1.051	.8835	1.135	1.084	1.045	2.394	1.094	1.112	1.140	1.030	1.183	3.092
	.778	1.001	1.270	.872	1.578	1.984	.808	.860	.968	.779	1.394	2.479
	.941	.747	1.212	.900	1.266	1.989	.850	.935	1.042	.836	1.263	2.612
		.893			1.178							
Total	2.770	3.524	3.617	2.856	5.067	6.37	2.752	2.907	3.150	2.645	3.840	8.183
Average	.923	.881	1.206	.952	1.267	2.123	.917	.969	1.050	.882	1.280	2.728

greater than the variance due to experimental error, the null hypothesis cannot be accepted and the lesser order interactions must be examined.

The findings of the analysis of variance are summarized in Table 2 and Figure 3. The conclusion drawn from the table and graphs was that a higher rainfall intensity, soil surface slope, and rainfall kinetic energy increased the depth of the mixing zone. However, the interaction that occurred between these parameters was quite complex. The rainfall intensity-kinetic energy interaction was significant only at the largest of the three slopes. The greater surface slope did show an increase of the mixing depth, but apparently the 0 percent slope damped out any effects induced by the rainfall intensity and energy.

Table 2. Summary of Results of Analysis of Variance

Source	Significance
ixHxS	significant
HxS	at i_1 - significant
	at i_2 - significant
ixS	at H_1 - only S was significant
	at H_2 - the interaction was significant
ixH	at S_1 - neither H nor i were significant
	at S_2 - only H was significant
	at S_3 - the Hxi interaction was significant

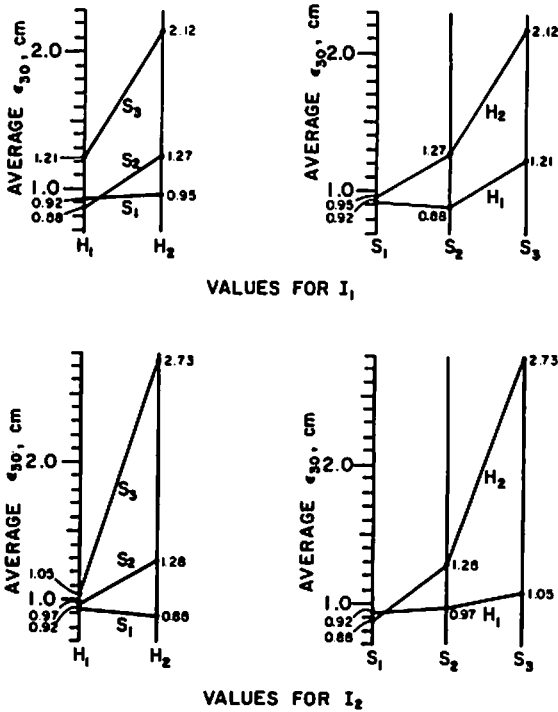


Figure 3. The relationship between mixing zone depth and the rainfall intensity, soil surface slope, and rainfall kinetic energy. (Note: the higher subscripts correspond to larger parameters values.)

A possible explanation for the results of the analysis of variance may be advanced by considering the work of Hutchler (1970). He shows that all of the raindrop energy is transmitted into the soil at zero depths of overland flow because no splash occurs that dissipates the energy. As the depth of the surface water increases, he found that as much as 95 percent of the raindrop's energy could be dissipated by splash. Therefore, with a lesser surface depth at steeper slopes, more energy is transmitted into the soil. Also, for the same depth of surface water, greater raindrop energy leads to more energy dissipation in the soil.

MATHEMATICAL MODEL ANALYSIS

The effects of the hydrologic variables on chemical transport was further examined by solving for the unknown term, ϵ_{50}^{cm} , in the mathematical model [Eq. (5) and (6)] after the other terms $\frac{\partial C}{\partial t}$ were estimated from experimental data. Three of these terms ($\frac{\partial C}{\partial t}$, $\frac{\partial C}{\partial x}$, and C) were

estimated by sampling the concentration of CaSO_4 in the overland flow at the downstream boundary and at the midpoint of the flume at discrete time points. Thus, values of $C(x,t)$ were available at the grid points shown in Figure 4.

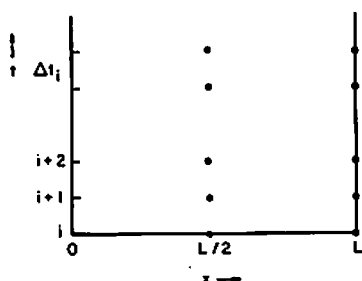


Figure 4. Domain of measurements.

The finite difference approximation of the partial derivatives of concentration were made as follows:

$$\left(\frac{\partial C}{\partial t}\right)^{i+1/2} = \frac{1/2(C_{L/2}^{i+1} + C_L^{i+1}) - 1/2(C_{L/2}^i + C_L^i)}{\frac{3}{4}L} \quad (11)$$

and

$$\left(\frac{\partial C}{\partial x}\right)^{i+1/2} = \frac{1/2(C^{i+1} + C^i)_L - (C^{i+1} + C^i)_{L/2}}{\frac{3}{4}L} \quad (12)$$

The coefficient C was approximated at $C_{\frac{3}{4}L}^{i+1/2}$ as follows:

$$(C)_{\frac{3}{4}L}^{i+1/2} = \frac{(C^{i+1} + C^i)_L + (C^{i+1} + C^i)_{L/2}}{4} \quad (13)$$

In these equations (11 - 13), the superscript i refers to time and the subscript L refers to distance.

Assuming a steady-state condition, the value of uh was obtained by the following identity:

$$uh = rx.$$

To obtain the value of h , the kinematic wave equation was used for positive slopes. The equation given by Eagleson (1970) for the height, h , is:

$$h = \left(\frac{rx}{\alpha}\right)^{1/m} \quad (14)$$

in which h = depth of overland flow (L)
 x = distance from the upstream boundary (L)

r = rainfall intensity (L/T)
 m is a parameter and α is a coefficient with units dependent on m .

Setting $m = 5$, which is an appropriate value for the disturbed laminar flow of this study, the parameter α can be estimated by equating the theoretical steady-state storage of water on the surface to a measured volume. The surface volume was calculated by integrating the recession hydrograph.

The theoretical surface storage can be obtained by integrating Eq. (14) between $x=0$ and $x=L_0$. The following relationship for α can be obtained from the integral expression:

$$\alpha = \frac{r L_0^{1+m}}{V^m \left(\frac{1}{m} + 1\right)^m} \quad (15)$$

where V is the volume of water on the surface at steady state.

The downstream boundary depth, h , for zero slopes can be assumed to be critical with

$$h = h_c \quad (16)$$

and

$$h_c = \left(\frac{q^2}{g}\right)^{1/3} \quad (17)$$

where h_c = critical depth of flow (L)
 q^2 = Discharge per unit width ($L^2 T^{-1}$)
 and g = acceleration due to gravity ($L T^{-2}$).

An expression for $\epsilon \frac{\partial C_m}{\partial t}$, can be obtained by eliminating the term $q_i(C_m - C)$ in Eq. (6) by substituting the relationship of Eq. (5). This can be done because with an initially saturated soil and a zero rate of infiltration q_0 equals q_i . The resulting equation is:

$$h \frac{\partial C}{\partial t} + uh \frac{\partial C}{\partial x} + \epsilon \frac{\partial C_m}{\partial t} = - Cr \quad (18)$$

Concentration data from five experimental runs were used to estimate the terms of Eq. (18). The resultant term $\epsilon \frac{\partial C_m}{\partial t}$, was then used to estimate C_m at various times. Because $\frac{dC_m}{dt}$ is needed for this determination of C_m at alternate times, the estimated value for ϵ after a 60-minute run time (using Eq. 9) was divided into the values of $\frac{dC_m}{dt}$. This gave values for $\frac{\partial C_m}{\partial t}$ at various times of the experiment except at time zero. To obtain this value, Eq. (18) was altered to the following form:

$$\frac{dC_m}{dt} = - \frac{Cr}{\epsilon} - \frac{rt}{\epsilon} \frac{dC}{dt} \quad (19)$$

This equation holds for the rising limb of the hydrograph because $h = rt$ and $\frac{\partial C}{\partial x} = 0$ at this time; therefore, at time zero, Eq. (19) becomes

WATERSHED MANAGEMENT

$$\frac{dC_m}{dt} = - \frac{Cr}{\epsilon} \quad (20)$$

Now having the values for $\frac{dC_m}{dt}$ at various times and the initial concentration of C_m , the values of C_m as a function of time can be obtained by numerical integration.

Figures 5, 6 and 7 show the resultant values of C_m plotted with C . It is apparent that C_m and C are not equal as some transport models assume (Crawford, et. al., 1973; Donigian, et. al., 1976; Steenhuis, et. al., 1978); however, it should be emphasized that C_m in these graphs is biased because of the method used in estimating ϵ . The estimate of ϵ is low because it was computed with the assumption that C_m is equal to C ; therefore, if one had a more realistic value of C_m (a value that should be greater than C) to use in Eq. (9), the new estimate for ϵ would be larger, thus making the difference between C_m and C more pronounced than in these figures.

Table 3 compares the calculated values of C_m at 30 minutes in Figure 7 (Run 53) with the actual concentration of CaSO_4 in the pore water in these samples. This comparison shows significant differences. The calculated concentration of C_m at 30 minutes in Figure 7 is 0.032 pph, whereas the average concentration in the upper 2 cm of soil is considerably higher. This difference may be due to the biased estimate of ϵ as previously discussed. However, the CaSO_4 concentration in the pore water also varies with depth and flow length which may be due to the greater dissipation of the raindrop energy within the surface runoff film at greater overland flow depths.

Table 3. Measured concentration of CaSO_4 in pore water of mixing zone for run 53.

Sample location	concentration, pph		
	Upstream Boundary	Mid-Section	Downstream Boundary
Surface Scratch	.016	.030	.058
.4 inch (1 cm) depth	.065	.088	.10
.4 - .8 inch (1-2 cm) depth	.18	.20	.16
Overland flow concentration		Mid-Section .074	Outflow .016

Saturated concentration equals .21 gm of CaSO_4 per 100 gm of distilled water.

Values of the exchange rate q_0 were solved using Eq. (5) with the collected and computed data for the runs presented in Figure 5, 6, and 7. The values of q_0 were found to have a slight variation with time. This variation may also be due to the biased estimate of ϵ from which the values of C_m used in equation (5) were computed. However, taking note of the average q 's for these runs (0.025, 0.017, 0.019, 0.022, and

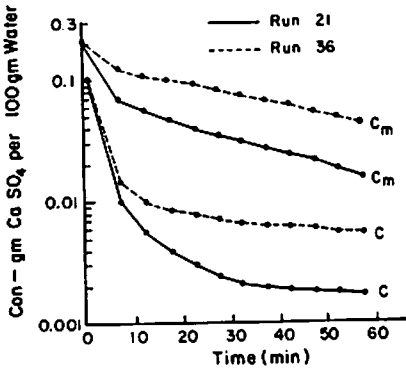


Figure 5. Relationship of C and C_m between Run 20 (i_2, H_1, S_2) and Run 36 (i_2, H_2, S_2).

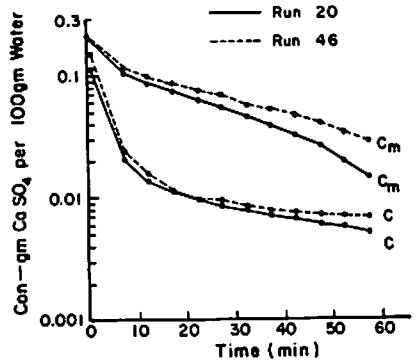


Figure 6. Relationship of C and C_m between Run 20 (i_1, H_1, S_2) and Run 46 (i_1, H_2, S_2).

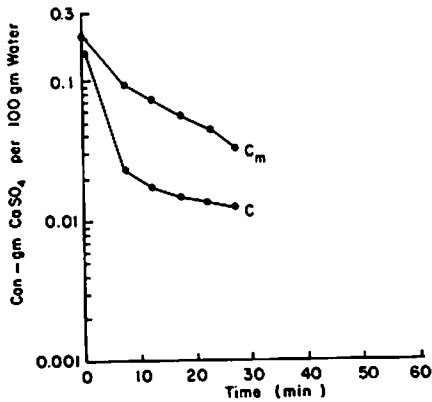


Figure 7. Relationship of C and C_m for Run 53 (i_1, H_2, S_2).

0.039 cm/min for runs 20, 21, 36, 46, and 53 respectively), there seems to be no consistent relationship between exchange rate and height of fall. The nearly equivalent exchange rates for the two intensities with the other parameters (slope and height of fall) held constant, supports the nonsignificance of the intensity factor in the statistical analysis. An increase of rainfall intensity would be expected to increase the exchange rate in the natural system. This is due to the correlation between the mean drop size and rainfall intensity in natural rainfall (Wischmeier and Smith, 1958). With an increase of the drop size there is a corresponding increase in kinetic energy. However, in this study the drop size was held constant for both intensities and its kinetic energy was varied by changing the height of fall.

SUMMARY AND CONCLUSIONS

In this study we examined the effects of rainfall intensity, soil surface slope, and rainfall kinetic energy on the release of a chemical from the soil into overland flow. Because of the small size of the experimental facility, with its short length of flow, this study more properly describes such transfer from interrill areas in the absence of sediment transport. The experimental rainfall kinetic energy was much less than that of natural rainfall. Because kinetic energy seems to have a significant effect on chemical transfer, this difference must be considered in interpreting the results of our study. The study demonstrated the following:

- 1) The concentration of a chemical species in overland flow is much lower than in the pore water. Thus, the assumption of an instantaneous equilibrium between the pore water and the overland flow is incorrect.
- 2) Release rates of a chemical species into the overland flow and the total transport in a given time were affected by kinetic energy and soil surface slope for the same rainfall intensity.
- 3) The scoop sample data showed that in the absence of infiltration, the chemical concentration profile in the soil pore water is affected by rainfall intensity and kinetic energy and by soil surface slope, thus showing that a concentration profile with leaching present should also account for the hydrologic parameters. Frere et al. (1975) neglected these hydrologic parameters when they assumed the runoff water to have the same chemical species concentration as that calculated for the pore water at the soil surface.
- 4) The statistical analysis showed that an increase of the magnitude of the hydrologic parameters increased the depth of the mixing zone. Also, the interaction between these parameters was quite complex.

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