

Solute and sediment transport at laboratory and field scale: Contributions of J.-Y. Parlange

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[1] We explore selected aspects of J.-Y. Parlange's contributions to hydrological transport of solutes and sediments, including both the laboratory and field scales. At the laboratory scale, he provided numerous approximations for solute transport accounting for effects of boundary conditions, linear and nonlinear reactions, and means to determine relevant parameters. Theory was extended to the field scale with, on the one hand, the effect of varying surface boundary conditions and, on the other, effects of soil structure heterogeneity. Soil erosion modeling, focusing on the Hairsine-Rose model, was considered in several papers. His main results, which provide highly usable approximations for grain-size class dependent sediment transport and deposition, are described. The connection between solute in the soil and that in overland flow was also investigated by Parlange. His theory on exchange of solutes between these two compartments, and subsequent movement, is presented. Both deterministic and stochastic approaches were considered, with application to microbial transport. Beyond contaminant transport, Parlange's fundamental contributions to the movement of solutes in hypersaline natural environments provided accurate predictions of vapor and liquid movement in desert, agricultural, and anthropogenic fresh-saline interfaces in porous media, providing the foundation for this area of research.

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1. Introduction

[2] Mass transport is a core factor in the analysis and prediction of environmental quality, for example, as a control on time scales of environmental system resilience. Apart from quantifying key elements of environmental system response, models of fate and transport are central to contaminant data analysis, risk assessment, and prognostic modeling,

to name but a few. Diffuse environmental pollution is ubiquitous [e.g., Carey *et al.* 2013; Islam and Tanaka, 2004; Novotny, 1999, 2007; Posen *et al.*, 2011]; thus techniques for environmental protection and remediation rely on the accuracy of models that predict outcomes of alternative strategies for remediation. It is no surprise then that modeling approaches are heavily embedded in analysis of transport processes. For example, a search within the 5500 papers published in *Water Resources Research* from 2000 to the present revealed that two-thirds include “model” in the title, abstract, or keywords. Nearly one in five papers includes both “model” and “transport” in these search categories.

[3] J.-Y. Parlange has made a vast array of contributions to environmental mass transport. Here, we focus on solute and sediment transport leaving, for example, his extensive work on water flow to be described by Assouline [2013]. Later, we explore his contributions to mass transport in overland flow (including sediment transport and transfers to flow from the surface soil) and in the near subsurface. Additionally, we briefly examine his contributions to thermodynamics of soil solutions. Our objectives are, first, to provide a guide to his body of work in this domain and, second, to give a flavor of his approach, which is both theoretical and physically based. Table 1 is intended to satisfy the first objective. The second objective is addressed in the following sections.

2. Column-Scale Solute Transport

[4] Column-scale solute transport is described by the classical advection-dispersion equation (ADE) [e.g., Barry, 1990; Bear, 1972]:

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Table 1. Summary of Parlange's Contributions on Solute and Sediment Transport

Topic	Section in Paper	Papers	Brief Description
Laboratory scale columns: Experiments, theory and analysis	2	<i>Parlange and Starr</i> [1975]	Showed that breakthrough curves from finite columns can be predicted by the solution for a semi-infinite column for $Pe > 4$.
		<i>Starr and Parlange</i> [1975]	Developed an approximate analytical approach for determining solute dispersion coefficient and (nonlinear) reaction rate.
		<i>Starr and Parlange</i> [1976a]	Presents an optimized method for determining the overall spatially-dependent transformation kinetics in a soil column experiment. The method was used to analyze a denitrification experiment.
		<i>Starr and Parlange</i> [1976b]	Experiments and modeling of stable and unstable displacement experiments.
		<i>Starr et al.</i> [1976]	Experiments describing cation exchange of radioactive tracers in soil column experiments, along with a simplified analysis.
		<i>Starr and Parlange</i> [1977]	Presented, modeled, and analyzed experimental data on tailing in breakthrough curves due to flow variations in the inlet porous plate of a soil column.
		<i>Parlange and Starr</i> [1978]	Analytical approximations for the 1-D transport equation, taking account of zero- and first-order kinetics.
		<i>Starr et al.</i> [1979]	Developed theory for determination of the effective diffusion coefficient for a solute undergoing sorption in a capillary tube and porous medium. The theory was validated using laboratory experimental data.
		<i>Starr and Parlange</i> [1979]	Presented and analyzed (using a simplified analytical model) soil column data on the snow-plow effect, which occurs when a high-concentration influent displaces a low-concentration initial solution in a porous medium.
		<i>Starr and Parlange</i> [1980]	Discussion on determining dispersion coefficients.
		<i>Starr et al.</i> [1980]	Reports extensive experiments and modeling of nitrogen transformations in soil.
		<i>Parlange et al.</i> [1982]	Developed approximations for the 1-D transport equation for a solute undergoing zero-order kinetics.
		<i>Starr et al.</i> [1982]	Presents experimental data and an analytical approximation for the precursor effect, which occurs during cation exchange experiments when a low-concentration influent solution displaces a high-concentration initial solution in a soil.
		<i>Barry et al.</i> [1983b]	Numerical modeling and model-based analysis of the snow-plow effect.
		<i>Parlange et al.</i> [1984]	Analytical approximation for the 1-D transport equation for arbitrary degradation kinetics.
		<i>Parlange et al.</i> [1985]	Discussion of the effect of the exit condition applied in modeling solute transport in a soil column.
		<i>Barry et al.</i> [1986]	Solves the 1-D transport equation via an interpolation method.
		<i>Barry et al.</i> [1987a]	Moment analysis of solute transport through layered media
		<i>Barry et al.</i> [1987b]	Numerical modeling and model-based analysis of the precursor effect.
		<i>Parlange et al.</i> [1992]	General relationship between resident and flux concentrations within and at the exit of a soil column. Zero- and first-order kinetics considered, as well as non-reactive tracers.
<i>Barry et al.</i> [1993]	Analytical approximation for 1-D solute transport with an arbitrary degradation reaction.		
<i>Xiong et al.</i> [2005]	Experiments on competitive sorption of metal cations in recycled wastewater to soil.		
Field scale transport	3	<i>Starr et al.</i> [1978]	Field study (two experiments) on vertical transport of water and chloride across soil layers.
		<i>Dayananda et al.</i> [1980]	Downward movement of a solute with water, with the latter modeled based on the field capacity.
		<i>Rose et al.</i> [1980]	Solute transport in a field profile—range of models, including water movement.
		<i>Barry et al.</i> [1983a]	General theory of 1-D vertical transport of water and solute, neglecting variability of hydraulic conductivity.
		<i>Barry et al.</i> [1985]	Modeling of transport of chloride and nitrate in a field soil with multiple fertilizer applications.
		<i>Starr et al.</i> [1986]	3-D experiments on water and solute movement in the field, demonstrating formation of fingers.
		<i>Parlange et al.</i> [1990].	Application of Miller scaling [<i>Miller and Miller</i> , 1956] to prediction of finger characteristics, with validation.
		<i>Nijssen et al.</i> [1991]	Test of a 1-D preferential flow and transport model using data from experiments on undisturbed soil cores.
		<i>Parlange et al.</i> [1991]	Unsaturated flow in a hillslope modeled using a linearly interpolated hydraulic conduction function.

Table 1. (continued)

Topic	Section in Paper	Papers	Brief Description
Soil erosion	4	<i>Stagnitti et al.</i> [1991]	Hillslope flow and transport model, accounting for pore-size distribution, surface runoff, evaporation, and precipitation.
		<i>Steenhuis et al.</i> [1991]	Downward, 1-D preferential flow and transport model and field application, accounting for hydraulic conductivity variability.
		<i>Selker et al.</i> [1992].	Experimental validation of 2-D and 3-D flow instability theory, showing that fingers are frequent and persistent features of vadose zone flow.
		<i>Stagnitti et al.</i> [1995]	Pesticide transport and biodegradation model combined with preferential/matrix flow theory and comparison with experiments.
		<i>Parlange et al.</i> [1996]	Preferential flow and transport based on pore-group sizes.
		<i>Griffioen et al.</i> [1998]	Characterization and dimensional analysis of published experiments on two-region solute transport.
		<i>Selker et al.</i> [1996]	Overview of unstable and preferential flow, from the perspective of predicting chemical transport in the field.
		<i>Stagnitti et al.</i> [1998]	Series of experimental validations of flow and transport theory under different field conditions.
		<i>Wallach and Parlange</i> [1998]	Two-region model concept applied to solute transport in a crack in a porous rock matrix.
		<i>Wallach et al.</i> [1998]	Systematic analysis of preferential water flow and solute transport from single fractures to multipore group models.
		<i>Steenhuis et al.</i> [2000]	Simplified solute transport model for preferential flow, including two model validations.
		<i>Wallach and Parlange</i> [2000]	Solute transport model for fracture flow with matrix exchange.
		<i>Stagnitti et al.</i> [2001]	Solute transport in undisturbed soil columns analyzed using single- and two-region models.
		<i>Parlange et al.</i> [2002b]	Explored differences in theories of unstable flow in Hele-Shaw cells and porous media.
		<i>Stagnitti et al.</i> [2003]	Preferential water flow model, with solute transport characterized by leaching distribution index.
		<i>Kim et al.</i> [2005]	Generalized preferential flow model, with solute transport, tested with laboratory data.
		<i>Sander et al.</i> [1996]	Analytical solutions for sediment individual size classes determined assuming the event is only time dependent.
		<i>Lisle et al.</i> [1998]	The Rose model generalized as stochastic Markov model considering soil particles alternating between rest and ejection states.
		<i>Hairsine et al.</i> [1999]	Solutions to the HR model in unsteady conditions provided.
		<i>Parlange et al.</i> [1999]	Analytical solutions of the HR model by exploiting short and long terms behavior determined.
		<i>Heilig et al.</i> [2001]	The HR model tested using laboratory flume experiment and evidence development of the shield layer.
		<i>Siepel et al.</i> [2002]	The HR model modified and tested taking different vegetative cover.
		<i>Hogarth et al.</i> [2004b]	Spatial and temporal solutions provided for dynamic change of the soil erosion due to the rainfall impact.
		<i>Rose et al.</i> [2007]	Impacts of ponding water depth and soil detachability on soil erosion tested and the HR theories validated using experimental data.
		<i>Sander et al.</i> [2007]	HR model theory tested successfully using published experimental data under net erosion and net deposition conditions.
		<i>Walker et al.</i> [2007]	Influence of infiltration on soil erosion processes investigated experimentally.
<i>Shaw et al.</i> [2008]	The stochastic form of Rose model tested experimentally.		
<i>Tromp-van Meerveld et al.</i> [2008]	Effect of sediment settling velocity on soil erosion delivery investigated under different experimental conditions.		
<i>Barry et al.</i> [2010]	Exact solutions of the HR model (assuming a single grain size), which are valid for all space and time, presented using appropriate assumptions.		
<i>Jomaa et al.</i> [2010]	Dependency of the boundary and initial conditions on rain splash studied with laboratory flume experiments.		
<i>Jomaa et al.</i> [2012a]	Effects of rock fragments on soil erosion delivery investigated and proportionality between soil erosion and area exposed to raindrops tested.		
<i>Jomaa et al.</i> [2012b]	HR model adjusted taking the rock fragments cover into account and tested using experimental data.		
<i>Jomaa et al.</i> [2013]	Antecedent soil conditions' effects on soil erosion investigated and the HR model tested through multiple rainfall events.		

Table 1. (continued)

Topic	Section in Paper	Papers	Brief Description
Solute exchange with surface water	5	<i>Walter et al.</i> [2001]	Integrated raindrop-driven transport of solutes from the mixing layer into surface runoff, diffusion-driven transport from deeper soil layers into the mixing layer, and infiltration.
		<i>Gao et al.</i> [2004]	Model of chemical transfer to overland flow, no parameter calibration needed.
		<i>Gao et al.</i> [2005]	Models combined sediment and chemical transport in overland flow, following the HR approach.
Pathogen transport	5	<i>Brush et al.</i> [1999]	Described <i>Cryptosporidium</i> transport in overland flow and soil columns.
		<i>Darnault et al.</i> [2003]	Experiments on preferential flow-driven transport of <i>Cryptosporidium parvum</i> oocysts.
		<i>Darnault et al.</i> [2004]	Prediction of <i>Cryptosporidium parvum</i> oocyst transport in preferential flow.
Thermodynamics of salt solutions	6	<i>Yeghiazarian et al.</i> [2006]	Stochastic Markov model of microorganism transport.
		<i>Parlange</i> [1973]	Analytical model for salt, liquid, and vapor phase water movement adjacent to a concentrated salt boundary.
		<i>Burns et al.</i> [2006a]	Thermodynamic potentials for saline solutions in variably saturated porous media.
		<i>Burns et al.</i> [2006b]	Effects of sodium chloride on retention and conduction of water in variably, saturated porous media.
		<i>Burns et al.</i> [2007]	Physically-based correction of the Buckingham-Darcy Law for flow of high strength salts in variably saturated porous media.

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

where c is solute concentration, D is the diffusion/dispersion coefficient, v is the advection velocity, z is position, and t is time. In equation (1), tracer transport is assumed. This model, with associated boundary conditions, is widely used to describe one-dimensional solute transport in homogeneous soil columns. For different circumstances, various modifications to equation (1) are possible, some of which will be addressed in subsequent sections.

2.1. Boundary Conditions

[5] Insights on the physical basis of boundary conditions used in obtaining solutions to equation (1) go back more than 50 years. It is not our purpose to present a detailed discussion of this important issue, rather to present boundary conditions appropriate for different conceptualizations.

[6] Solute concentrations can be measured in two different physical circumstances. If a liquid sample is withdrawn from a medium quickly, then the concentration measurement is called a resident concentration, c_r . More precisely, this means that the characteristic dimension of the space from which the liquid is withdrawn, L_s , is much greater than the distance moved by the pore fluid over the sampling time, t_s , i.e., $L_s \gg vt_s$. The other circumstance is where liquid is collected as it crosses a surface, typically the column exit. In this case, the concentrations measured are flux-weighted, and so are termed flux concentrations, c_f . The relationship between these two measurement types is given by [Kreft, 1981; Kreft and Zuber, 1978; Parker and van Genuchten, 1984; Sposito and Barry, 1987]:

$$c_f = c_r - \frac{D}{v} \frac{\partial c_r}{\partial z}. \quad (2)$$

[7] Both c_r and c_f satisfy (1). As noted by Parlange et al. [1992], a zero- or first-order sink term added to the right

side of equation (1) leaves it unchanged under equation (2) [Kreft and Zuber, 1986].

[8] Because of the different physical interpretations of c_r and c_f , different boundary conditions can apply in determining solutions to equation (1), depending on the measurement technique and the setup of the column. Solute transport experiments in soil columns involve saturated or, less often, uniformly unsaturated steady flow. For saturated flow, solute enters via a surface (or basal) reservoir and drains into a reservoir or tube. Apparatus-induced dispersion occurs if the water flow is nonuniform, due to curvilinear flow paths within the column [Barry, 2009; Greiner et al., 1997]. For the surface reservoir ($z = 0$), the boundary condition is [Hulburt, 1944]:

$$c_f(0, t) = c_0, \quad (3)$$

where c_0 is the concentration in the reservoir. The corresponding condition for c_r follows from equation (2) [Bastian and Lapidus, 1956; Wehner and Wilhelm, 1956, 1958]:

$$c_0 = c_r - \frac{D}{v} \frac{\partial c_r}{\partial z}, \quad z = 0. \quad (4)$$

[9] In laboratory experiments, inflow into a soil column is often through a porous plate, which can contain preferential flow paths. Starr and Parlange [1977] observed that these paths are a mechanism for producing tailing in solute breakthrough curves in short columns (Figure 1). Within the porous plate, the possibility for solute exchange with immobile regions was examined, and an analytical approach was developed to account for this exchange. Using laboratory experiments, Starr and Parlange [1977] showed that even if uniform flow was established within a small distance of the entrance plate, the effects of nonuniformities propagate in the soil column. This occurs because

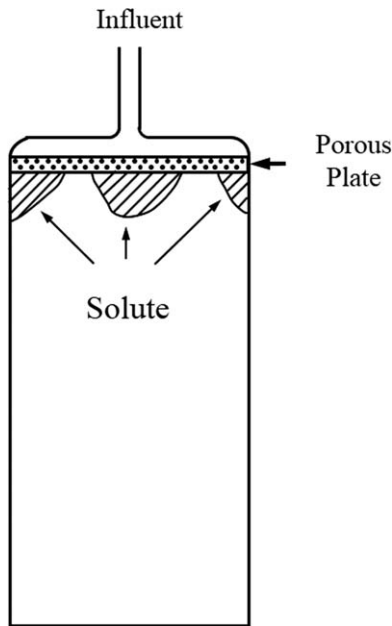


Figure 1. Schematic of nonuniform flow occurring in a soil column experiment as a result of imperfections in the overlying porous plate (after *Starr and Parlange* [1977] used with permission).

smoothing of disturbances by transverse diffusion/dispersion must take place, and so the effluent concentrations will exhibit apparatus-induced dispersion.

[10] To model the effect of the nonuniform flow field, *Starr and Parlange* [1977] replaced the nonuniform flow regions shown in Figure 1 by a mixed zone, below which the flow is 1-D (one-dimensional), characterized by a mixing length (which is an adjustable parameter). Figure 2 shows a comparison between their model results and experimental data. Their single-parameter mixing model clearly describes well the breakthrough curve tailing induced by the nonuniform flow at the column entrance.

[11] For the case of a column that drains into a tube or reservoir, the exit condition at $z = L$ (the column length) is [*Barry and Anderson*, 1996b; *Brenner*, 1962; *Danckwerts*, 1953]:

$$\frac{\partial c_r(L, t)}{\partial z} = 0. \quad (5)$$

[12] For a free-draining column, c_r is identically zero [*Barry and Anderson*, 1996a; *Barry and Sposito*, 1988]; however, equation (5) is the most common case in practice. Solutions to (1) are much simpler if (5) is applied as $z \rightarrow \infty$ rather than at $z = L$, i.e., the column is treated as being semi-infinite (as was the case for the model results in Figure 1). *Parker* [1984] suggested that a macroscopic discontinuity can occur at the exit boundary in a soil with large, continuous pores, or fractures, near the exit, since then concentrations within the column are unaffected by the outflow boundary. For this situation, the semi-infinite model is more apt [*Parker*, 1984]. Because the soil structure near the column exit is usually not known a priori, *Parlange et al.* [1992] considered that the semi-infinite column or (5)

defined a range of possible exit boundary conditions, with the lowest value of c_r at the boundary given by the semi-infinite case, and the maximum when (5) is used. Another approach is to model a soil column as a two-layer medium (i.e., the exit apparatus is modeled as a layer with different transport properties), as investigated in various studies including *Shamir and Harleman* [1967], *Barry and Parker* [1987], *Barry et al.* [1987a], *Leij and Van Genuchten* [1995], and *Schwartz et al.* [1999].

[13] Although not connected to this physical interpretation, substitution of equation (5) into equation (2) gives $c_r(L, t) = c_f(L, t)$, which leaves a corresponding mathematical ambiguity in terms of finding solutions to equation (1). It does, however, suggest that solutions for c_r (for a finite column) and c_f (for a semi-infinite column) could coincide at $z = L$, as observed by *Gershon and Nir* [1969]. This finding was examined by *Parlange and Starr* [1975], who showed that the different solutions were essentially identical at $z = L$ for column Péclet numbers, $Pe = vL/D$, greater than about four (see section 2.2.1).

[14] The effect of the boundary condition at $z = L$ extends within the soil column over a length scale of order D/v [*Parlange et al.*, 1985], so the Péclet number condition of *Parlange and Starr* [1975] was based on the assumption that the effect of the boundary condition at $z = L$ does not interact with the condition applied at the column entrance. Their approach was also exploited to develop analytical approximations and to improve numerical solutions [*Bajracharya and Barry*, 1993, 1994; *Barry et al.*, 1986; *Parlange et al.*, 1982, 1985; *Parlange and Starr*, 1975, 1978]. Because boundary conditions can have such impact on the quantification of laboratory and similar experiments, their interpretation in different circumstances has a significant history and contemporary interest [*Coronado et al.*, 2007, 2009; *Gimmi and Fluhler*, 1998; *Novakowski*, 1992a, 1992b; *Peters and Smith*, 2001; *Schwartz et al.*, 1999].

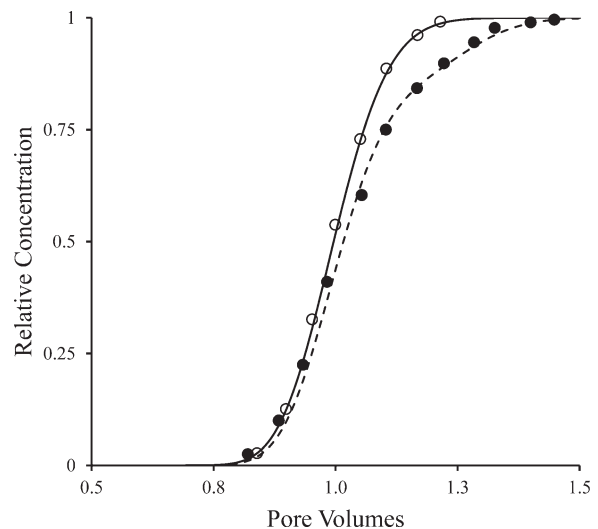


Figure 2. Laboratory column breakthrough curves and model predictions. Open (solid) circles, breakthrough concentrations without (with) plate-induced tailing. The lines are model predictions. This figure is reproduced (with permission) from *Starr and Parlange* [1977], where details of the experiments and modeling approach are provided.

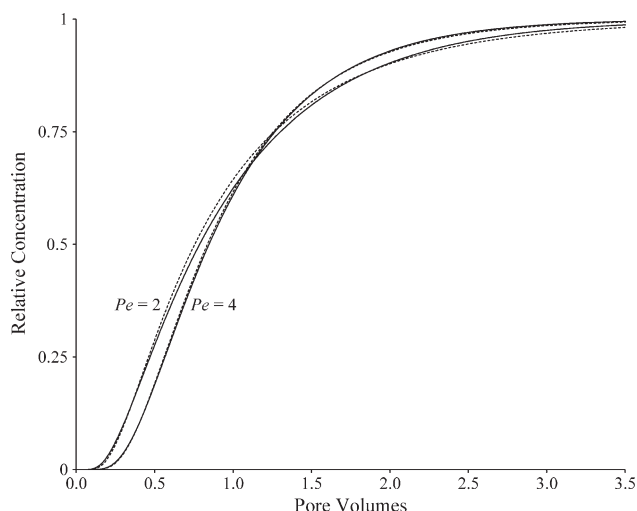


Figure 3. Solute concentrations at the end of a laboratory soil column. The approximation (dashes) from equations (8) and (9) is compared with the exact solution (line), equations (6) and (7), for two values of Pe .

2.2. Approximate Solutions for Finite Soil Columns

2.2.1. Solute Concentration at the Exit of a Laboratory Column

[15] Solutions to equation (1) for different boundary conditions can rapidly become complex and unwieldy to compute. For finite soil columns, solutions to equation (1) take the form of infinite series [van Genuchten and Alves, 1982], which could be prone to errors during numerical evaluation.

[16] As previously noted, Parlange and Starr [1975] explored the conditions under which resident and flux concentration solutions effectively coincide at the column exit. They also analyzed parameters determined from using simplified analytical expressions. Specifically, they presented an analytical approximation to equations (1), (4), and (5), given by Brenner [1962] and evaluated at $x = L$:

$$\frac{c(L, t)}{c_0} = 1 - 16Pe \sum_{i=1}^{\infty} \beta_i \exp\left(\frac{Pe}{2} - \frac{v^2 t}{4D} - \frac{\beta_i^2 Dt}{L^2}\right) \times \frac{2\beta_i \cos(\beta_i) + Pe \sin(\beta_i)}{(4\beta_i^2 + Pe^2)(4\beta_i^2 + Pe^2 + 4Pe)}, \quad (6)$$

where the β_i are the positive roots of:

$$Pe[4\beta_i \cot(\beta_i) + Pe] = 4\beta_i. \quad (7)$$

[17] If equations (6) and (7) give the correct evolution of concentration at the outflow end of a soil column, then this can be approximated by the semi-infinite solution satisfying equations (1), (3), and (5) (with $L \rightarrow \infty$ in the latter) [Lapidus and Amundson, 1952]:

$$\frac{c(L, t)}{c_0} = \frac{1}{2} \left[\operatorname{erfc}\left(\frac{L - vt}{\sqrt{4Dt}}\right) + \exp(Pe) \operatorname{erfc}\left(\frac{L + vt}{\sqrt{4Dt}}\right) \right]. \quad (8)$$

[18] Parlange and Starr [1975] found that if equation (8) is used to compute the apparent dispersion coefficient (D_{ap})

in a laboratory column experiment, then the true dispersion coefficient, D , is related to D_{ap} by:

$$Pe = Pe_{ap} - 1, \quad (9)$$

where $Pe_{ap} = vL/D_{ap}$. In Figure 3, results from equations (6) and (7) are compared with equations (8) and (9). Note that this comparison has not been made previously, as Parlange and Starr [1975] used their approximation to equations (6) and (7) in their comparison. For $Pe = 2$, there are some differences evident between the two solutions. These differences are much diminished for $Pe = 4$ and reduce further for $Pe > 4$ [Parlange and Starr, 1975] (results not shown).

2.2.2. Interpolation Between Resident and Flux Concentrations at a Column Exit

[19] In the foregoing subsection, no distinction was made between resident and flux concentrations. Depending on how concentrations are measured, both are possible. Furthermore, the soil structure at the outlet adds uncertainty concerning the concentration measured [Parker, 1984]. A brief description of the methodology employed by Parlange in several papers to approximate the range of possible measurements is now presented. The culmination of this approach is found in Parlange *et al.* [1992], which tackled the question of the ambiguity in exit boundary condition.

[20] As already indicated, Parker [1984] observed that a discontinuity in c_r at $z = L$ "should occur in fractured or aggregated porous media having continuous relatively large pores." Motivated by this observation, Parlange *et al.* [1992] considered two limiting cases identified by Scheidegger [1957], one being that dispersion in the soil column is due to transverse diffusion (typically between mobile/immobile regions), and the second being that dispersion is due to mixing of pore-scale flow paths. For the first case, the semi-infinite solution given by equation (8) (with L replaced by z and c replaced by c_f) can be used on the assumption that the boundary at $z = L$ does not induce any back-diffusion within the soil column. The second case induces concentration changes upstream from the boundary, and equation (5) applies.

[21] Starting from equation (8), Parlange *et al.* [1992] provided an analytical approximation that accounts for the possible behaviors of concentrations measured in the breakthrough curve, i.e.,

$$\frac{c_r}{c_0} = \frac{c_f}{c_0} + \lambda \exp\left(\frac{vz}{D}\right) [f(L) - f(z)], \quad 0 \leq z \leq L, \quad (10)$$

where c_f is given by equation (8) and

$$f(z) = \left\{ \operatorname{erfc}\left(\frac{z + vt}{\sqrt{4Dt}}\right) \left[1 + \frac{v}{2D}(z + vt) \right] - v \sqrt{\frac{t}{D\pi}} \exp\left[-\frac{(z + vt)^2}{4Dt}\right] \right\}, \quad (11)$$

with the interpolation parameter λ ($0 \leq \lambda \leq 1$) defined by:

$$\frac{\partial c_r(t, L; \lambda)}{\partial z} = (1 - \lambda) \frac{\partial c_r(t, L; 0)}{\partial z}. \quad (12)$$

[22] If $\lambda = 1$, then equation (10) is an approximation satisfying the boundary condition equation (5); the case of

back-diffusion at the column exit. The other limit, $\lambda = 0$, corresponds to the case where back-diffusion is negligible, and the column behaves as if it were semi-infinite. If fitted to experimental data at the column exit ($z = L$), then equation (9), which relates the true Pe to the fitted value (Pe_{ap}), is replaced by:

$$Pe = Pe_{ap} - \lambda. \quad (13)$$

[23] The approach sketched here was extended by Parlange *et al.* [1992] to include the cases of zeroth and first-order reactions in the governing transport equation (1).

3. Field-Scale Solute Transport

[24] At the field scale, water movement is non-steady and non-uniform. For the latter, heterogeneity in soil properties results in variability in water movement even when the water flux through the soil surface is uniform. Vertical transport of water parcels in the soil profile was shown to be approximately log-normal for steady water input (due to the hydraulic conductivity distribution), so this distribution has been used in field-scale models [Biggar and Nielsen, 1976; Jury, 1982; Nielsen *et al.*, 1973]. Concerning the variability of soil properties, Nielsen *et al.* [1973] noted that “seemingly uniform land areas manifest large variations in hydraulic conductivity values” and so “our ability to make predictions over a large area from a single plot can range from good to unsatisfactory.” In this vein, Jury [1982] took the “pessimistic point of view that . . . spatial variability of water and solute transport . . . renders measurement of the hydraulic and retention parameters of a field soil all but impossible.” This situation has not changed in the more than 30 years that have passed if one considers the challenge of obtaining spatially resolved measurements of these parameters. For this reason, approaches based on simple concepts of water movement due to changes in volumetric moisture content are still valuable in practical circumstances.

[25] The approach presented here summarizes a group of papers where further details and insights are available [Barry *et al.*, 1983a; Dayananda *et al.*, 1980; Raats, 1975, 1977; Rose *et al.*, 1982b, 1982c; Rose and Parlange, 1982]. First, water flow is considered, then solute transport.

3.1. Simplified Approach to Vadose Zone Water Flow at the Field Scale

[26] Field scale water movement is a notoriously difficult problem if detailed quantification is desired. This is due to the strong nonlinearity of vadose zone water movement and variable soil hydraulic properties. In addition, field measurement of boundary conditions (e.g., evaporation) is likewise challenging. Nonetheless, predictive models are essential, which led Parlange and colleagues to expand on a simple mass-balance theory.

[27] The water flow model is derived from the following simplified picture: Inputs and outputs affecting water movement in the vadose zone are net water flux (infiltration–surface evaporation) at the soil surface and plant uptake within the soil profile. Except on short time scales (which were not considered), the maximum water content within the root zone is field capacity, θ_{fc} , i.e., if the water content is greater than field capacity, downward motion of the excess water

must occur. In the plant root zone, water removal by plant uptake reduces the moisture content below θ_{fc} . Similarly, if evaporation exceeds infiltration in a given time period, then water is removed from the soil profile. Such a simplified picture ignores the detailed dynamics of water flow. Rather, it assumes a time scale over which rapid water redistribution can be ignored, e.g., a day or a week.

[28] In this modeling framework, the soil profile is divided into the root zone (depth D_R), from which plant water uptake can occur, and the lower soil profile, which is assumed to be always at field capacity. The average moisture content in the root zone, θ , varies according to:

$$\theta(t) - \theta_{fc} = \int_{t_0}^t \frac{I(\bar{t}) - E(\bar{t})}{D_R(\bar{t})} d\bar{t}, \quad (14)$$

where I is the water flux at the soil surface, E is the evapotranspiration rate due to plants, D_R is the plant rooting depth (usually taken as constant), and t_0 is an arbitrary initial time at which $\theta = \theta_{fc}$. Equation (14) assumes that water removal by plants is uniform in the root zone. To be clear, water enters the soil through the surface at rate I (which can be negative), whereas it is removed uniformly from the soil profile by plants at rate E . In formal terms, the right side of equation (14) should include a Heaviside step function to reduce the change in θ to zero when $\theta \geq \theta_{fc}$. In calculations, this condition simply indicates periods when water movement below the root zone occurs. That is, movement of water below the rooting depth occurs via piston flow whenever excess water is drained below D_R .

[29] We next consider the position of a water front in the soil profile. If water enters the soil profile at time t_0 , then its front position, z_f , at time t is found by solving:

$$\theta_J \frac{dz}{dt} = \frac{I - zB}{D_R}, \quad z < D_R, \quad (15)$$

where θ_J is given by equation (14) and $B = I$ if $\theta < \theta_{fc}$ or $B = E$ otherwise. A detailed derivation of (15) is given by Dayananda *et al.* [1980]. In Figure 4, the underlying mass balance leading to (15) is shown schematically. An underlying physical assumption is that the water front can move downward only. As mentioned, evaporation removes water from the whole root zone but does not change the position of the water front. Thus, downward movement of the water front occurs whenever water infiltrates into the soil. This approach permits tracking of the position of water in the soil profile for arbitrary input conditions. If water infiltrates into the soil at time t_0 , at time $t \geq t_0$ its front position is given by the solution to equation (15):

$$z_f(t) = \exp\left(-\int_{t_0}^t \frac{B}{D_R \theta_J} d\bar{t}\right) \int_{t_0}^t \frac{I}{\theta_J} \exp\left(\int_{t_0}^{\bar{t}} \frac{B}{D_R \theta_J} d\bar{t}\right) d\bar{t}, \quad (16)$$

$$z_f < D_R.$$

[30] Some insight into the behavior of $z_f(t)$ can be readily obtained for the simple case where the soil profile is initially at θ_{fc} , the rooting depth D_R is constant and the net flux of water through the soil surface is exactly balanced by

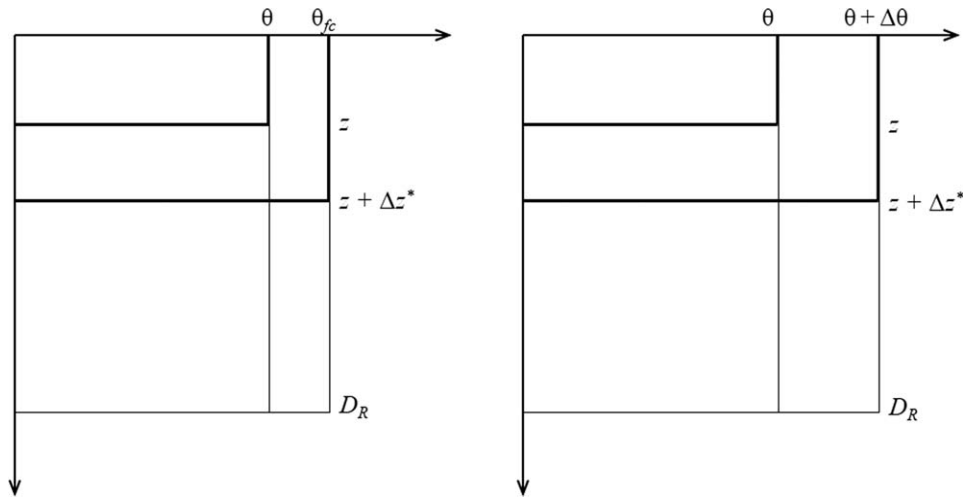


Figure 4. Water movement after an input of $I\Delta t$ of rainfall/irrigation to a soil profile for which the initial moisture content is θ , where $\theta < \theta_{fc}$. In both cases, D_R is the rooting depth, which defines the region from which evaporation can occur, and the water front is at position z prior to water addition. For the left-hand case, the added water is sufficient to raise the moisture content to θ_{fc} . The excess water moves the initial water front position from z to $z + \Delta z^*$, which is determined from mass balance. In the right-hand case, the added water is insufficient to raise the water content to θ_{fc} . In this case, the moisture content increases to $\theta + \Delta\theta$, while the water initially at depth z moves to depth $z + \Delta z^*$. The rate of movement of the water front at z in both cases is given by equation (15) (after *Dayananda et al.* [1980], used with permission).

water removal by plants from the soil profile, i.e., $I = E$. For these conditions, in equation (15) $\theta_J = \theta_{fc}$ and $B = E$. For $t_0 = 0$, equation (16) reduces to:

$$\frac{z_f(t)}{D_R} = 1 - \exp\left(-\frac{Et}{D_R\theta_{fc}}\right), \quad z_f < D_R. \quad (17)$$

[31] In equation (17), the water front is initially at the soil surface and eventually reaches the root zone depth (D_R) as $t \rightarrow \infty$. The water front moves downwards initially due to the imbalance between water influx at the surface and removal of water throughout the entire depth, D_R . It cannot penetrate below the root zone since, as it approaches depth D_R , any water entering the profile is removed via evaporation and so there is no longer any imbalance at the location z_f .

[32] To summarize, the soil profile is divided into two parts, the root zone where the moisture content has a maximum value of θ_{fc} , and the profile below the root zone where the moisture content is always at θ_{fc} . The position of an infiltration front is given by equation (16) in the root zone. Below the root zone, since from equation (14) the amount of water leaving the root zone is known, the water front position is calculated from the piston flow assumption.

3.2. Solute Movement During Stable Flow

[33] Darcy flow is the basis of advection-dispersion transport theory, applied to our knowledge first to the field by *van der Molen* [1956] to predict the rate of desalinization of the Dutch polder soils after inundation by the sea. He derived the ADE from chromatography theory, based on the assumption that all water percolating through the soil moves approximately with the same velocity as predicted by the flux obtained from Darcy's law divided by the fraction of volume occupied by mobile water. The solute disperses

around the solute front that moves with the average velocity and is described by a dispersion coefficient. It is generally assumed based on the implications of creeping flow (as required by Darcy's law) that the dispersion coefficient varies linearly with the average solute velocity [*Gelhar et al.*, 1992; *Jury et al.*, 1991]. Subsequently, the ADE was tested with repacked [*Brush et al.*, 1999; *Huang et al.*, 1995; *Wierenga and van Genuchten*, 1989] and undisturbed soil columns [*Mohammadi and Vancloster*, 2011].

[34] We consider the simple case of a nonreactive (and nondecaying) tracer that enters the soil at a known concentration and is passively taken up by the plants along with water. Because the theory in section 3.1 permits tracking of water fronts, for the circumstances considered here, the solute concentration in water that enters the soil at some time t_0 moves to $z_f(t)$ at time $t \geq t_0$, where $z_f(t)$ is given by equation (16). Note that, as t_0 is arbitrary, this approach gives the position of any water "front" of interest. In other words, water that enters the soil at a specified time is located some time later at a position, z_f . Note that it is assumed here that infiltrating water displaces all water that is in the profile.

[35] Dispersion of solute at field scales can be significant. Inclusion of dispersion in the solute transport equation and using the theory in section 3.1 does not yield a model amenable to analytical solution. However, analytical results are easily obtained if the solute advection rate, v , is given by the dz_f/dt , calculated from (15). Then, (1) becomes:

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial z^2} - \frac{dz_f}{dt} \frac{\partial c}{\partial z}. \quad (18)$$

[36] Equation (18) was solved for arbitrary boundary and initial conditions on a semi-infinite domain by *Barry and*

Sposito [1989]. For the practical case where the dispersion coefficient is proportional to the advection rate, i.e., $D = \chi dz_f/dt$ ($\chi > 0$ is the dispersivity) [Bear, 1979], the solution is straightforward since then it involves a simple temporal rescaling. For example, for this case, the solution for the boundary condition $c(0, t) = c_0$, the solution is:

$$\frac{c(z, t)}{c_0} = \frac{1}{2} \left[\operatorname{erfc} \left(\frac{z - z_f}{\sqrt{4\chi z_f}} \right) + \exp \left(\frac{z}{\chi} \right) \operatorname{erfc} \left(\frac{z + z_f}{\sqrt{4\chi z_f}} \right) \right]. \quad (19)$$

[37] Successful applications of this approach were presented by *Rose et al.* [1982a, 1982b], who simulated field data of *Saffigna et al.* [1977] and *Chichester and Smith* [1978].

3.3. Solute Movement During Preferential and Unstable Flow

[38] Preferential flow in the vadose zone refers to several phenomena that have in common the nonuniform and often rapid movement of water, dissolved solutes, and adsorbed chemicals (to colloids). This rapid movement bypasses the bulk of the soil matrix, reducing the potential for pollutant adsorption and/or degradation and increasing the threat of groundwater and surface water contamination. Preferential and unstable flow is not restricted to the subsoil but can also be seen where water moves over a surface. Amongst other effects, it results in the formation of rills and gullies in eroding landscapes and can be even noted as the streaks (“tears”) of a film of wine draining on the inside surface of a glass.

[39] Preferential flow was described first by *Lawes et al.* [1882] during field drainage experiments in which they noted that the soil drained through macropores initially from all parts of the profile. This is in contrast to the traditional view based on Darcy flow where water flow moves as slug with a speed that is averaged over all pores (the case considered in section 3.2). Darcy’s approach (i.e., averaged conditions) continued to be used over the next century because the simplifying assumptions of isotropic homogeneous soils were convenient as calculations could be dealt with more easily. However, the discovery of pesticide contamination of Long Island aquifers in the early 1980s made it clear that under field conditions groundwater contamination by toxic chemicals could not be explained in all cases by the usual application of the ADE because a small fraction of the pesticides moved much faster to the groundwater than the average speed of the water. As pesticide concentrations, for example, are toxic at concentrations in the parts per billion level, a small fraction (usually less than 0.1% of that applied) can raise concentrations in the groundwater above the drinking water standard.

[40] The Long Island findings led to a surge in preferential flow research. The term “preferential flow” has more than 2200 citations in the last 30 years (Science Citation Index). Early work showed that preferential water and solute flow could be separated into three distinct categories: First, macropore flow in well-structured soils [*Beven and Germann*, 1982; *Lawes et al.*, 1882; *Quisenberry and Phillips*, 1976]; second, fingered (or column flow), manifested as unstable wetting fronts in granular soils with the total flux of water many times smaller than the saturated conduc-

tivity [*Culligan et al.*, 1997, 2002; *Hill and Parlange*, 1972; *Parlange and Hill*, 1976; *Parlange et al.*, 2002b; *Raats*, 1975]. In the latter category, we can also place unstable flows in water-repellant soils [*Bauters et al.*, 1998; *Dekker and Ritsema*, 1994a, 1994b, 1995; *Hendrickx et al.*, 1993; *Ritsema and Dekker*, 1994, 1995; *van Dam et al.*, 1990]. Third, there is funnel flow, in which water and solute flows in the finer grained soils overlying coarse layers [*Kung*, 1990a, 1990b]. As noted by *Dekker and Ritsema* [1994b], preferential flow is more the rule than the exception.

[41] Despite the various forms of preferential flow, there are some general behaviors that can be elucidated. The flow patterns for the different types of preferential flow are nearly always the same for both fingered flow in sandy or water-repellent soils and macropore flow in structured soils. Under low flow conditions, as shown by *Hendrickx et al.* [1993] and *DiCarlo et al.* [1999] for homogeneous sandy soils, and *Mohammadi and Vanclouster* [2011] for undisturbed structured soil cores, the difference in speeds is small and the ADE can be used to describe the solute movement in the soil, implying that the traditional Darcy’s law approach (as used, for example, in section 3.2) is valid. When flow increases, water and solute can move preferentially. Dye patterns indicate how water and solutes move through the soil. At the soil surface, water infiltrates uniformly and then flows through this upper horizontal layer in mostly preferential vertical paths. This is equivalent to a stationary wetting front. The horizontal layer (distribution zone) varies in thickness from a few millimeters to the thickness of the plow layer of approximately 30 cm. The amount of solute moving out of this horizontal layer or mixing layer can be described as follows for steady state flux:

$$c = \frac{M}{W} \exp \left(-\frac{It}{W} \right), \quad (20)$$

where M is mass per unit area applied and W is the apparent water depth in the distribution zone:

$$W = d(\theta_d + \rho_b). \quad (21)$$

[42] Here, d is the depth of the distribution zone, k is the adsorption partition coefficient, ρ_b is bulk density, and θ_d is volumetric moisture content in the distribution zone.

[43] The formation of preferential flow paths below the mixing zone depends on the imposed flux. These are shown schematically in Figure 5 as fingers or preferred flow paths. For wetter conditions, structured and sandy soils behave differently to water-repellent soils. For structured soils, the greater the flux (and the wetter the soil), the faster the chemicals move downward. This occurs because for wetter soil there is less interaction with the matrix and solute moves at an increased rate through the largest pores. Finger mergers can also occur at subtle changes in soil texture, as these changes affect the local hydraulic conductivity [*Kung et al.*, 2000a, 2000b].

[44] The geometrical characteristics of fingers that develop from unstable flow are also determined by soil texture and the imposed water flux. However, the influence of the latter is much less than the former [*Parlange et al.*, 1990; *Selker et al.*, 1992]. Figure 6 shows finger diameters

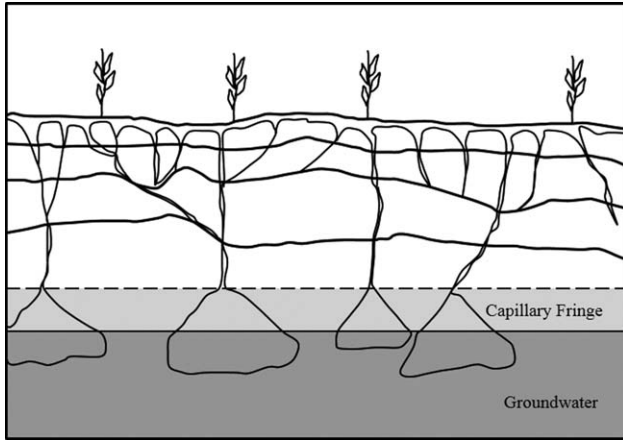


Figure 5. Different mechanisms lead to unstable and preferential flow in the vadose zone. These locations of these localized flow paths are unknown a priori, as are the locations of textural interfaces that lead to mergers (after Selker et al. [1996] used with permission).

for different soil textures, with little variation due to the total downward water flux. Specifically, for unstable finger flow in an initially dry sandy soil the downward speed of the chemicals does not increase when the imposed flux is increased. The velocity, V , is calculated from:

$$V = \frac{K_s}{\theta_s} \left(1 + \frac{\psi}{L_b} \right), \quad (22)$$

where K_s is the saturated hydraulic conductivity, θ_s is the saturated volumetric moisture content, and ψ is the matric potential in the finger behind the wetted bulb (which has length L_b). From conservation of mass, the integral of the speed and area that takes part in transport equals the imposed flux for steady-state application. Thus, the proportion of the soil that is wet (i.e., transmits fingers), A_w , can be expressed as [Darnault et al., 2003, 2004; Kim et al., 2005; Selker et al., 1996]:

$$A_w = \frac{I}{V\theta_s}. \quad (23)$$

[45] For sandy soils, this means that the velocity of the front is independent of the imposed flux when prior fingers do not exist. Selker et al. [1996] determined that in field soils the storm with maximum rainfall intensity determines the maximum proportion of the soil that is wet, $A_{w,max}$. As long as the soil does not dry out, inflow from any subsequent storms will flow through this wetted area. Thus, for steady-state conditions the average downward solute velocity is:

$$V = \frac{I}{A_{w,max}\theta_f}, \quad (24)$$

where θ_f is the moisture content in the finger.

[46] This principle was demonstrated by Kim et al. [2005]. In their experiment, water and solutes were infiltrated first at a low flow rate of 0.4 cm h^{-1} . In the second cycle, the high application rate was around 1.7 cm h^{-1} and

in the third cycle 0.4 cm h^{-1} . The breakthrough time for the first and second cycles was approximately the same while for the third cycle the time for breakthrough was the longest.

[47] Finally, assuming that the advective-dispersive flow in the finger itself with the distribution layer at the surface and the exponential boundary condition (20), the concentration in the finger can be written as [Kim et al., 2005; Toride et al., 1995]:

$$\frac{c(z,t)}{c_0} = \frac{1}{2} \exp\left(-\frac{It}{W}\right) \left\{ \exp\left[\frac{vz}{2D}(1-\alpha)\right] \text{erfc}\left(\frac{z-vt\alpha}{\sqrt{4Dt}}\right) + \exp\left[\frac{vz}{2D}(1+\alpha)\right] \text{erfc}\left(\frac{z+vt\alpha}{\sqrt{4Dt}}\right) \right\}, \quad (25)$$

where

$$\alpha = \sqrt{1 - \frac{4DI}{WV^2}}. \quad (26)$$

[48] Equation (25) is just a solution to equation (1) for the case of an exponentially decaying surface condition in a semi-infinite spatial domain [Marino, 1974; van Genuchten and Alves, 1982].

[49] Although counterintuitive in structured soils, water arrives earlier than for sandy soils. Similarly, Nimmo [2007] noted that experiments for which a continuous relatively high flux was applied, the geometric mean speed was $13 \pm 6 \text{ m d}^{-1}$. Speeds in cracks were above the mean while those in the surrounding soil were below.

[50] Much progress has been made in modeling solute transport during preferential flow. One of earliest was the dual porosity model in which the ADE is modified to include a mobile region and an exchange coefficient with the stagnant regions [Coats and Smith, 1964]. Other preferential flow models are those of Ahuja et al. [1993], Faybishenko et al. [2000], Gaudet et al. [1977], Griffioen and Barry [1999], Kung et al. [2000a, 2000b], Ritsema and Dekker [1994], Ritsema et al. [1998], and Steenhuis et al. [1994b]. Jury and co-workers took a different approach by formulating the transfer function model [Javaux and

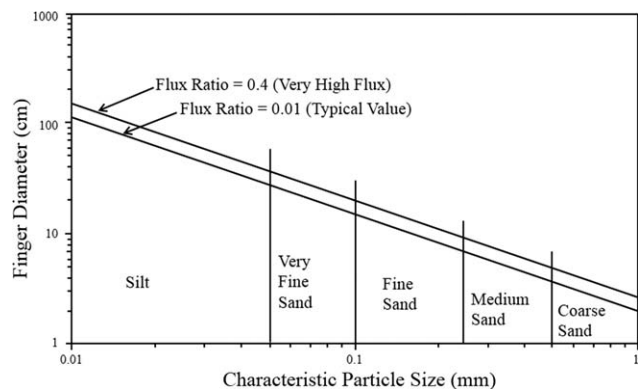


Figure 6. Diameter of unstable fingers for different soil water fluxes as they vary with soil texture (after Selker et al. [1996] used with permission). The flux ratio is the average water flux relative to the saturated hydraulic conductivity.

Vanclooster, 2003; *Jury et al.*, 1990; *Jury and Roth*, 1990; *Roth and Jury*, 1993]. In this approach, the solute flow input response at a certain depth is calculated from the solute flow input response in the layer above when the correlation is known between the points [*Nissen et al.*, 2000]. Other models that have the capacity to include preferential flow are RZWQM [*Ahuja et al.*, 1991], MACRO [*Larsbo and Jarvis*, 2003; *Moeys et al.*, 2012], and PEARL [*Tiktak et al.*, 2012].

[51] The model performance for practical applications is limited because, in all cases, the input parameters cannot always be determined a priori. Although many studies have been carried out with blue dye characterizing preferential flow paths, the problem is that most of these studies have been carried out under ponded conditions to find the greatest number of preferential flow paths. Pesticide leaching occurs, however, under natural rainfall. In this case, in structured soils only a fraction of the stained pores have a role in the transport. Although recently a function for determining the macroporosity was established [*Jarvis et al.*, 2009], it is not possible to predict what paths will take part in the transport, making it difficult to predict leaching of contaminants precisely. Because of this, in practice simple models do as well as the more complicated models.

4. Sediment Transport in Overland Flow: The Hairsine-Rose Model

[52] While the physical processes controlling the erosion and transport of sediment are different to the transport of solutes in overland flow (Section 5) or through porous media, there are strong mathematical similarities between the governing equations. Hence it is not surprising that Parlange has also made significant contributions to the understanding of erosion mechanisms through the development of simple and accurate analytical approximations to the flow equations. In this section, we provide a brief overview of the environmental importance of sediment transport modeling, the governing equations, and the physical processes involved. In particular, we focus on Parlange's contributions to this field through his work on the Hairsine-Rose (HR) model [*Hairsine and Rose*, 1991, 1992a, 1992b; *Rose et al.*, 1983a, 1983b].

[53] The transport of eroded material from land to surface water bodies by overland flow is an important environmental problem, promoting the eutrophication of surface waters, damaging freshwater ecosystems, and causing the contamination of surface waters. Sediment derived from the soil is itself a pollutant. It reduces light penetration and degrades freshwater ecosystems and is a carrier of pollutants such as pesticides, fertilizers, and pathogens. The development and spatial extent of severe hypoxic coastal zones is known to be caused by rivers discharging increased levels of sediment-sorbed nutrients originating from agricultural runoff. Hypoxic zones commonly occur throughout the year in the Black and East China Seas, Baltic Sea, and in the Gulf of Mexico [*Boesch et al.*, 2009; *Diaz and Rosenberg*, 2008].

[54] Over the past 40 years there have been many physically based mathematical models developed that try to estimate or predict erosion rates. These have been applied across the different increasing spatial scales of laboratory,

plot, hillslope, and watershed with varying degrees of success [*Boardman*, 2006]. The most commonly used models are WEPP [*Flanagan and Nearing*, 1995], KINEROS2 [*Smith et al.*, 1995], LISEM [*de Roo et al.*, 1996], and EUROSEM [*Morgan et al.*, 1998]. These models are all based on the kinematic approximation for overland fluid flow and mass conservation for suspended sediment:

$$\frac{\partial h}{\partial t} + \frac{\partial q}{\partial x} = P - I, \quad (27)$$

and

$$\frac{\partial(hc^{sp})}{\partial t} + \frac{\partial(qc^{sp})}{\partial x} = G, \quad (28)$$

where P is the rainfall rate, c^{sp} is the suspended soil particle or sediment concentration, $q = n^{-1}\sqrt{S_0}h^{5/3}$ is volumetric flow per unit width, h is flow depth, S_0 is bedslope, n is Manning's friction coefficient, and G represents erosion source or sink terms. Nearly all models have G as being made up from two terms; one for soil detachment by raindrop impact, D_I , and the second for the net rate of soil detachment by the flow, D_F , hence:

$$G = D_I + D_F. \quad (29)$$

[55] Note that in equation (29) deposition of particles due to gravity is not included as a separate rate process; rather it is the net difference between the rates of flow detachment and the deposition that is determined through D_F . Hence, different formulations are used for D_F depending on whether sediment transport is occurring under net erosion ($D_F > 0$) or net depositional conditions ($D_F < 0$). To distinguish between the two requires the concept of a predefined transport capacity, T_c , which defines the maximum sediment flux, $q_s = qc^{sp}$, that the flow can transport. Thus, if $q_s < T_c$, more soil will be eroded, but if $q_s > T_c$ then the additional sediment will be deposited. Difficulties arise with the concept of transport capacity as it is well known that, even with the same flow rate, bedslope and soil type, T_c will be different between net erosion and net depositional conditions due to its dependence on the soil's particle size distribution [*Polyakov and Nearing*, 2003; *Sander et al.*, 2007]. Consequently, not only is T_c hysteretic, but the individual size class contributions are hysteretic also [*Sander et al.*, 2007, 2011], which makes prescribing a predetermined equation for T_c impractical.

[56] Over the past decade, there has been a greater recognition of the importance of the role of the particle size distribution in soil erosion [*Boardman*, 2006; *Govers*, 2011]. This is not only important in determining sediment fluxes across a landscape but also chemical and microbial fluxes due to the preferential binding of contaminants to clay and silt sized particles [*Morgan and Quinton*, 2001; *Schijven and Hassanizadeh*, 2000]. The HR model is unique in that it represents separately the three erosion mechanisms of rainfall detachment, runoff entrainment, and gravity deposition; hence it does not require the concept of transport capacity. It also describes the soil by its particle size probability density function and was the first to develop a fully multisize class model. In addition, the HR model

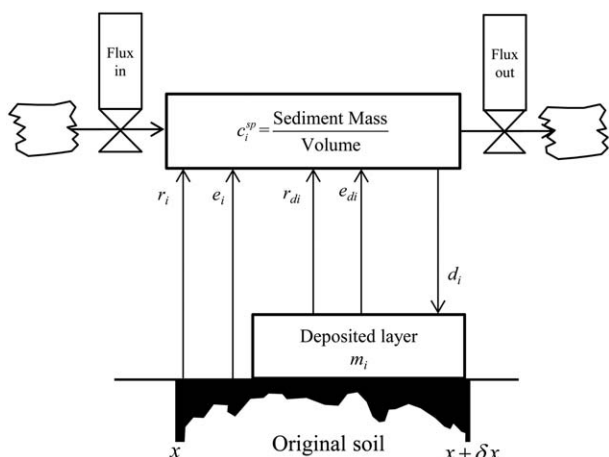


Figure 7. Conceptual layout of HR model (after *Hairsine and Rose* [1992b]). The original cohesive soil is suspended into the overland flow and deposited into a less-cohesive layer, which is in turn eroded.

recognizes that previously eroded cohesive soil depositing on the soil surface creates a covering layer of noncohesive sediment. Because of the size-selective nature of the deposition process, the distribution of size classes in this layer is different to that of the original soil. A simple but informative experiment conducted by *Heilig et al.* [2001] showed how the development and evolution of this deposited layer changes through time. Consequently, the HR model requires mass conservation equations of both suspended and deposited sediment for each size class. A conceptual layout of the HR model is shown in Figure 7, which results in the system of equations:

$$\frac{\partial(hc_i^{sp})}{\partial t} + \frac{\partial(qc_i^{sp})}{\partial x} = e_i + e_{di} + r_i + r_{di} - d_i, \quad i = 1, 2, \dots, N \quad (30)$$

and

$$\frac{\partial m_i}{\partial t} = d_i - r_{di} - e_{di} \quad i = 1, 2, \dots, N, \quad (31)$$

with

$$\begin{aligned} e_i &= a(h)p_iP(1-H), & e_{di} &= a_d(h)PH\frac{m_i}{m}, & r_i &= \frac{F_r}{J_e}p_i(1-H)(\Omega - \Omega_{cr}), \\ d_i &= \nu_i c_i^{sp}, & r_{di} &= \frac{F_r}{gh} \frac{\rho_s}{\rho_s - \rho} H(\Omega - \Omega_{cr}) \frac{m_i}{m}, & H &= \min\left(1, \frac{m}{m^*}\right), \\ \frac{a}{a_0} &= \frac{a_d}{a_{d0}} = \begin{cases} 1, & h \leq \bar{h} \\ (\bar{h}/h)^\delta, & h > \bar{h}. \end{cases} \end{aligned} \quad (32)$$

[57] In equations (30) and (31), i refers to a specific size class, N is the total number of size classes, e_i and e_{di} are the rainfall detachment rates from the original uneroded soil and deposited layer, respectively, r_i and r_{di} are the sediment entrainment rates (flow-driven detachment) from the uneroded soil and deposited layer, respectively, d_i is the deposition rate, c_i^{sp} is the suspended sediment concentration, $c^{sp} = \sum c_i^{sp}$ is to total sediment concentration, m_i is the

mass per unit area of sediment in the deposited layer, and $m = \sum m_i$. From equation (32), a and a_d are flow depth-dependent soil detachability coefficients for the original soil and deposited layer (with threshold values denoted by the subscript 0), respectively, \bar{h} is the threshold depth for the detachment rates, δ is a soil characteristic parameter, p_i ($0 < p_i \leq 1$ and $\sum p_i = 1$) is the proportion of sediment in size class i of the original uneroded soil, H ($0 \leq H \leq 1$) is the protection factor provided by the deposited layer, F_r is the fraction of excess stream power effective in entrainment, J_e is the specific energy of entrainment, ρ is the water density, ρ_s is the particle solid density, g is the magnitude of gravitational acceleration, $\Omega = \rho g S_0 q$ is the stream power with Ω_{cr} the critical threshold stream power below which r_i and r_{di} are zero, ν_i is fall velocity, and m^* is the mass per unit area of deposited sediment required to protect the original soil from further erosion. The HR concept of separating out the layer of deposited sediment has now been adopted in more recent models [*Kinnell*, 2005; *Nord and Esteves*, 2005].

[58] The deposited layer in the HR model has the same role as the immobile zone in mobile-immobile models of contaminant transport in groundwater [*Griffioen et al.*, 1998; *Li et al.*, 1994] or that of the transient storage zones in the OTIS model [*Runkel*, 1998] for contaminant transport in rivers. Thus, it retards the downslope movement of the sediment, resulting in the type of long tail breakthrough curves as shown in Figure 8 [*Heilig et al.*, 2001, 2006].

4.1. Analytical Approximations

[59] *Sander et al.* [1996] developed an approximate solution to equations (30) and (31) and applied it to data from the flume experiments of *Proffitt et al.* [1991]. In these experiments, rainfall detachment was the only erosive mechanism, i.e., $r_i = r_{di} = 0$. There was no infiltration into the soil, the flume surface was initially covered with a layer of water, and there was no discharge of water onto the flume at $x=0$. The approximate solution assumed that

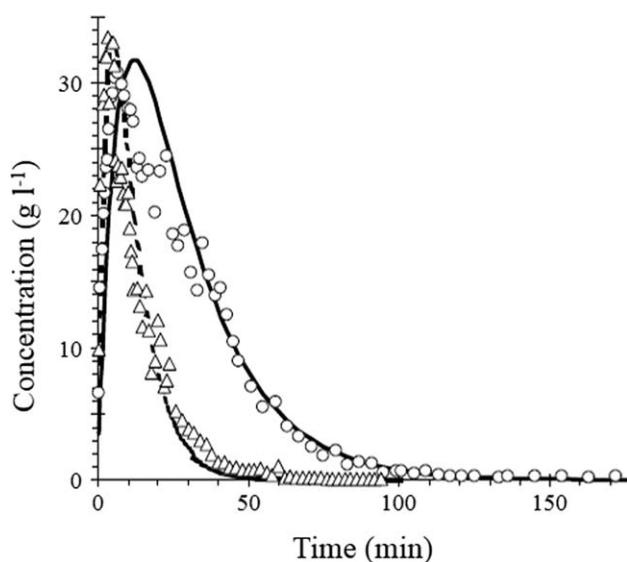


Figure 8. Measured and predicted clay concentrations as a function of time for $P = 35 \text{ mm h}^{-1}$ (circles) and $P = 100 \text{ mm h}^{-1}$ (triangles) (*Heilig et al.* [2001] used with permission).

spatial effects could be neglected and hydraulic conditions remained constant and was given in terms of eigenvalues and eigenvectors of the linear system of coupled ordinary differential equations (taking $p_i = 1/N$):

$$\frac{d}{dt}(hc_i^{sp} + m_i) = \frac{aP}{N}(1-H) - Pc_i^{sp}, \quad (33)$$

$$\frac{dm_i}{dt} = v_i c_i^{sp} - a_d P \frac{m_i}{m^*}, \quad (34)$$

$$t = 0, \quad c_i^{sp} = 0, \quad m_i = 0, \quad (35)$$

with

$$H = \frac{1}{m^*} \sum_{i=1}^N m_i. \quad (36)$$

[60] As solutions to equations (33) and (34) could only be obtained numerically by *Sander et al.* [1996] (because the eigenvalues were computed), very little information on the form and structure of the solution could be obtained. *Parlange et al.* [1999] derived an approximate analytical solution that gave a clear understanding of the physical processes controlling the transport. This was achieved by recognizing that the solution revolved around just two timescales, a short timescale dependent on rainfall impact and a much longer timescale concerned with the movement of particles by advection.

[61] For short timescales where $m^*/a_d h \ll Pt/h \ll 1$, the term $-Pc_i^{sp}$ in equation (33) can be neglected, thus it follows that one can write $hc_i^{sp} + m_i = s(t)$. When combined with equation (34) and integrated, this results in explicit formulas for the short-time behavior of the suspended sediment and the deposited masses as:

$$hc_i^{sp} = \left(1 + \frac{m^* v_i}{a_d h P}\right)^{-1} s, \quad (37)$$

$$m_i = \frac{v_i/P}{a_d h/m^* + v_i/P} s, \quad (38)$$

$$1 - \left(\sum_{i=1}^N \frac{v_i/P}{a_d h/m^* + v_i/P}\right) \frac{s}{m^*} = \exp\left(-\frac{1}{N} \sum_{i=1}^N \frac{v_i/P}{a_d h/m^* + v_i/P} \frac{aPt}{m^*}\right) \quad (39)$$

and

$$H(t) = 1 - \exp\left(-\frac{1}{N} \sum_{i=1}^N \frac{v_i/P}{a_d h/m^* + v_i/P} \frac{aPt}{m^*}\right). \quad (40)$$

[62] *Parlange et al.* [1999] noted that the long-time solution behavior is governed by the rates of deposition and detachment in the deposited layer being in balance to leading order. Physically, this means that while the contributions of the individual size classes to the deposited layer continue to change slowly, the total mass in the deposited layer remains almost constant, hence from equation (34) $v_i c_i^{sp} = a_d P m_i / m^*$. This permits the direct integration of

equation (33) and, following some additional simplifications, *Parlange et al.* [1999] obtained:

$$H = \frac{1-H}{N} \frac{a}{a_d} \sum_{i=1}^N \left[1 - \exp\left(-\frac{\frac{a_d P}{m^*} t}{\frac{a_d h}{m^*} + \frac{v_i}{P}}\right)\right] \frac{v_i}{P} + \sum_{i=1}^N \exp\left(-\frac{\frac{a_d P}{m^*} t}{\frac{a_d h}{m^*} + \frac{v_i}{P}}\right) \frac{m_{i0}}{m^*}, \quad (41)$$

$$\frac{m_i}{m^*} = \frac{v_i}{P} \frac{a}{a_d} \frac{1-H}{N} + \exp\left(-\frac{\frac{a_d P}{m^*} t}{\frac{a_d h}{m^*} + \frac{v_i}{P}}\right) \left(\frac{m_{i0}}{m^*} - \frac{v_i}{P} \frac{a}{a_d} \frac{1-H}{N}\right), \quad (42)$$

where m_{i0} is the value of m_i at the start of the long time solutions and given by:

$$m_{i0} = \frac{ahv_i}{P + \frac{m^*}{a_d h} v_i} \left(\sum_{i=1}^N \frac{P + \frac{a}{a_d} v_i}{\frac{a_d h P}{m^*} + v_i}\right)^{-1}. \quad (43)$$

[63] The level of agreement between these approximations and the numerical solution of *Sander et al.* [1996] is good, making the approximations usable in predicting and analyzing experimental data. The coupling between the size classes in equations (33) and (34) is through the deposited layer and, therefore, H . The important simplification of *Parlange's* analysis is that it allows a priori estimates of $H(t)$ to be obtained and to therefore decouple the system of $2N$ equations into N pairs of equations, one pair for each size class.

[64] *Proffitt et al.* [1991] also conducted experiments wherein the no-inflow boundary condition at $x=0$ was replaced with a constant discharge inflow. Under these conditions, the spatial dependence of the suspended sediment concentration cannot be neglected and the steady state spatially varying depth profile— $h^{5/3} = (h_0^{5/3} + Px)n/\sqrt{S_0}$ with ($h_0 = h$ at $x=0$)—must also be included. The resulting system is [*Hogarth et al.*, 2004a]:

$$h \frac{\partial c_i^{sp}}{\partial t} + q \frac{\partial c_i^{sp}}{\partial x} = \frac{aP}{N}(1-H) - Pc_i^{sp} - \frac{\partial m_i}{\partial t}, \quad (44)$$

$$\frac{\partial m_i}{\partial t} = v_i c_i^{sp} - \eta P m_i, \quad (45)$$

where $\eta = a_d/m^*$. Solutions to equations (44) and (45) for the initial conditions $c_i^{sp} = 0$, $m_i = 0$, and boundary condition $x=0$, $c_i^{sp} = 0$ in general are obtained numerically. However, being a first-order hyperbolic system the method of characteristics can be used in order to develop approximate analytical solutions. The solution has two branches with one branch close to steady state and controlled by the boundary condition, while the second branch is controlled by the initial condition and has limited spatial dependence. The position where these two branches meet is determined by the characteristic emanating from $x=0$ at $t=0$.

[65] By exploiting the different behavior in the two branches, a fully analytical approximation for $c_i^{sp}(x,t)$ can be found as [*Hogarth et al.*, 2004a]:

$$c_i^{sp}(x,t) = c_{i+}^{sp} - \frac{1}{2}(c_{i-}^{sp} - c_{i+}^{sp}) \operatorname{erfc}\left\{\frac{\ln[h_i^{-1}h(x)]}{\sqrt{4\omega_i t}}\right\}, \quad (46)$$

where

$$\omega_i = \frac{9\nu_i\eta P^3}{25(\eta Ph_i + \nu_i)^3}, \quad (47)$$

$$c_{i-}^{sp} = \frac{a}{N}(1 - H^-) \left\{ 1 - \left[\frac{h_0}{h(x)} \right]^{\frac{5}{3}} \right\} \quad (48)$$

and

$$c_{i+}^{sp} = \left[1 - \exp\left(\frac{-aPt}{Nm^*} \sum \frac{\nu_i}{\nu_i + \eta Ph}\right) \right] \times \left\{ \frac{a(1 - H^+)}{N} \left[1 - \exp\left(\frac{-Pt}{h + \nu_i(\eta P)^{-1}}\right) \right] + \frac{\eta P m^* \exp\left(\frac{-Pt}{h + \nu_i(\eta P)^{-1}}\right)}{(\nu_i + \eta Ph) \sum \frac{\nu_i + a^{-1}\eta P m^*}{\nu_i + \eta Ph}} \right\}. \quad (49)$$

[66] Because the second branch has negligible spatial dependence then H^+ can be found from equation (41) while H^- is found from:

$$\left(\frac{h}{h_0}\right)^{\frac{5}{3}} \left(\frac{\sum \nu_i}{\eta P m^* + 1}\right) = \frac{a \sum \nu_i}{a(1 - H^-) \sum \nu_i - N \eta P m^* H^-}. \quad (50)$$

[67] The position where the two branches meet is denoted as h_i , with $h_i^{5/3} = (h_0^{5/3} + Px_i)n/\sqrt{S_0}$ and is found by iteratively solving:

$$h_i - h_0 + \frac{\nu_i}{\eta P} \ln\left(\frac{h_i}{h_0}\right) = \frac{3}{5}Pt. \quad (51)$$

[68] Alternatively, the solution to equation (51) can be expressed explicitly using the Lambert W function [Barry et al., 1995a, 1995b, 2000, 2005; Corless et al., 1996; Parlange et al., 2002a] by solving ($W = \eta Ph_i/\nu_i$):

$$W \exp(W) = \frac{\eta P h_0}{\nu_i} \exp\left[-\frac{\eta P}{\nu_i} \left(h_0 + \frac{3}{5}Pt\right)\right]. \quad (52)$$

[69] Equation (51) shows that the meeting point not only varies through time but also that it is different for every size class. Hence, each size class moves downstream under its own speed and the approach to steady state is governed by the slowest moving particle size class. Hogarth et al. [2004a] showed that this characteristic speed is given by $q(h + \nu_i/\eta P)^{-1}$; hence all particles with fall velocities such that $\nu_i \ll \eta Ph$ will be carried along with the flow and effectively do not settle. This characteristic speed decreases as ν_i increases due to interchange between the particular size class being in suspension and in the deposited layer. A further implication of this result is that in order to estimate the total mass of sediment leaving the flume, the smallest size class should be chosen to satisfy $\nu_i \ll \eta Ph$. This analysis of Hogarth et al. [2004a] highlights the significant contributions that analytical techniques and approximate solutions

can make to understand not only the underlying structure of the solution but also the effect of parameter interactions on solution behavior.

[70] While the majority of the analytical approximations have been developed for rainfall-driven erosion only, Hogarth et al. [2011] considered the case where runoff-driven erosion and deposition processes occurred. By comparing their analytical results with those from a full numerical solution, they exploited the differences between the behavior of the smaller and larger particle sizes and showed how they contribute to the growth of the deposited layer. They found that the limiting steady state solution provided a good estimate for the long term contribution of the small particle sizes and that the limiting solution as $x \rightarrow \infty$ provided a good representation of the early rapid growth of H from the larger particle sizes. By providing an intermediate approximation that interpolated between these limits, Hogarth et al. [2011] obtained a solution for all x and t for all size classes with good accuracy.

4.2. Simple Experiments

4.2.1. Development of the Deposited Layer

[71] The solutions above show the importance of the deposited layer on erosion dynamics. What they also imply is that it is insufficient to collect typical data on the sediment flux at $x=L$ to validate the HR model fully; data on the formation of the deposited layer are also needed [Barry et al., 2010]. Parlange and coworkers demonstrated this with a series of simple experiments. The development of the deposited layer from suspended sediment concentrations was shown by Heilig et al. [2001]. Their experiment consisted of a square level surface (7 cm \times 7 cm) over which a constant shallow depth of water (5 mm) was subject to various rainfall rates. The soil was composed of two size classes being 10% clay and 90% sand. Thus, the soil can be regarded approximately as having two extreme behaviors in that the clay and sand have zero and infinite settling velocities, respectively. Physically this means that once clay particles become suspended they will then flow out of the domain and that the suspension time for sand particles is so small that it can be neglected. This leads to a simple solution of the HR model (without spatial dependence due to the experimental conditions) [Heilig et al., 2001]:

$$c_1^{sp} = \exp\left(\frac{-Pt}{h}\right) \frac{a}{10 - \frac{ah}{m^*}} \left\{ \exp\left[\left(10 - \frac{ah}{m^*}\right) \frac{Pt}{h}\right] - 1 \right\}, \quad (53)$$

and

$$H = 1 - \exp\left(-\frac{9aPt}{10m^*}\right). \quad (54)$$

[72] A prediction of this solution is that the peak concentration is independent of the rainfall rate. This prediction was confirmed by the experimental data (Figure 8) using rainfall rates that varied by a factor of three. This is an unusual finding in terms of what is commonly seen in the literature and it is partly as a result of the artificial soil type that was created for the experiment. For a soil containing a smooth distribution of size classes, this result no longer holds, although the dependence of the peak concentration

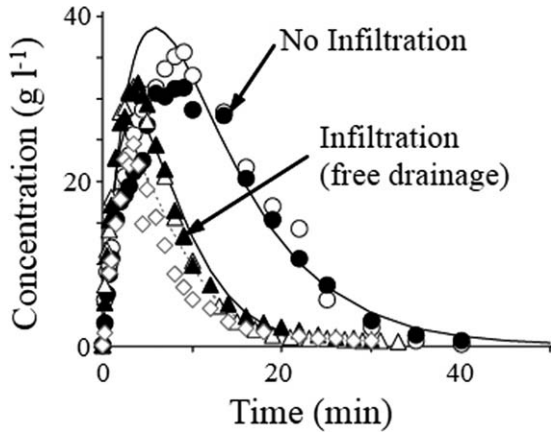


Figure 9. Experimental clay concentrations (symbols) and the predicted concentrations (lines). Circles = no infiltration, triangles = infiltration, diamonds = increased infiltration. Filled and open symbols are for replicate experiments (*Walker et al.* [2007] used with permission).

on rainfall rate is still quite mild if all other parameters remain the same.

[73] The HR model predicts the experimental data in Figure 8 as it models the development of the deposited layer shielding the underlying soil. Figure 5 of *Heilig et al.* [2001] demonstrates how this layer evolves through time and shows its impact on the erosion of the clay particles. Initially, there is a rapid rise in the clay concentration following the commencement of rainfall. As the shield then begins to grow, the raindrop access to the clay particles in the original soil is reduced and the concentration reaches a maximum. Over time, the deposited layer completely covers the original soil and the clay concentration eventually goes to zero. Note that the concentration going to zero is a consequence of this experimental setup and does not apply to traditional flume experiments. An increase in the rainfall rate also results in an increase in the rate of formation of the deposited layer such that the peak concentration remains the same. The change in rainfall rate predominantly affects the rate of decline in concentrations after the peak.

4.2.2. Infiltration Effects

[74] The use of simple experimental designs to elucidate physical processes was extended by *Walker et al.* [2007], who studied the impact of infiltration on rainfall-driven erosion. The same soil composition as *Heilig et al.* [2001] was used. Infiltration was established by allowing water to drain through the bottom of the soil container, thereby establishing a vertical velocity component to the overland flow that had the effect of increasing the settling velocity of all particles by the same amount [*Tromp-van Meerveld et al.*, 2008]. The results of their experiments are shown in Figure 9, which compares concentrations with and without infiltration. They found that infiltration has a significant impact of sediment loss resulting in a more rapid rise to a lower peak concentration and more rapid decline than in the absence of infiltration. They observed that the deposited layer not only formed faster (due to the increased settling velocity), but it was also thinner than with infiltration. *Walker et al.* [2007] also showed that the analytical solution of equations (53) and (54) applied to their experiments

although for a reduced deposited layer. A smaller m^* leads to a larger coefficient of t in the exponential terms in these equations resulting in a more rapid rise and faster decline as seen in the data. The reason why m^* became smaller was not fully understood, but potential explanations included a possible increase in the density of the deposited layer as a result of infiltration and/or greater raindrop energy was absorbed by the infiltration water and therefore less was available for detaching soil particles.

4.2.3. Ponding Depth

[75] In another set of experiments, *Gao et al.* [2003] looked at the impact of surface water depth on the mass of soil eroded or detached. To reduce the interactions between erosion processes so that just the role of flow depth on detachment could be studied, the previous theme of creating simple experiments was continued. By using a soil composed of clay particles, the deposition process could be neglected along with the formation of a deposited layer. Second, no water was allowed to escape from the experimental device through infiltration or overland flow and all of the rainfall contributed to the surface water depth, hence for a constant rainfall rate, P , the surface water depth was given by $h = Pt$. For these conditions they showed that the HR model simplifies to

$$\frac{d(hc^{sp})}{dt} = a(h)P, \quad (55)$$

with $a(h)$ given by equation (32). Defining the mass per unit area of suspended sediment as $m^{sp} = hc^{sp}$, then the solution of equation (55) for the initial condition of $m^{sp} = 0$, $t = 0$ is:

$$m^{sp} = a_0 h, \quad h \leq \bar{h} \quad (56)$$

and

$$m^{sp} = \begin{cases} a_0 \bar{h} + \frac{a_0}{1-\delta} \bar{h}^\delta (h^{1-\delta} - \bar{h}^{1-\delta}), & h \leq \bar{h}, \delta \neq 1, \\ a_0 \bar{h} + a_0 \bar{h} \ln(h/\bar{h}), & h > \bar{h}, \delta = 1. \end{cases} \quad (57)$$

[76] We see from equation (56) that for flow depths less than \bar{h} , m^{sp} increases linearly with t (as $h = Pt$), and so it is linearly dependent on the rainfall rate P . Both of these linear dependencies were confirmed by the experimental data [see Figure 6 of *Gao et al.*, 2003] across a range of rainfall rates between 6 and 43 mm h⁻¹ and for soils that were either initially saturated and unsaturated. Equation (57) was also shown to give an excellent match ($R^2 = 0.98$) to the measured data with $a_0 = 0.23$ g mm⁻¹, $\bar{h} = 8.9$ mm, and $\delta = 4$ for the saturated soil; and $a_0 = 0.038$ g mm⁻¹, $\bar{h} = 8.4$ mm, and $\delta = 4$ for the unsaturated soil. The lower values of detachability occur for the saturated soil as a result of reduced cohesion between the particles compared to an unsaturated soil.

[77] Overall, these three simple experiments and accompanying analytical solutions examined (i) the role of an evolving deposited layer, (ii) the impact of infiltration of soil detachment, and (iii) the effect of flow depth and rainfall rate on detachment. They demonstrated the physical applicability of the conceptual mechanistic process understanding on which the HR model is based. This body of

work on soil erosion typifies the style of scientific contributions that Parlange has made throughout his career in all fields that he has worked. That is, isolate the key fundamental physical processes and interactions that determine the system response, use this to guide which terms must be kept in the governing equations, and then proceed to derive straightforward, but accurate approximate solutions. Lastly, follow this up with well-designed experiments that justify and corroborate the basis of the physical simplifications used to develop his approximate solutions.

5. Transfer of Solutes From the Soil to Overland Flow

[78] Traditionally, two distinct approaches had been used in solute transport from soil into surface runoff: the lumped, mixing layer approach and the diffusion approach. The mixing layer approach assumes that rainwater, soil solution, and runoff water mix instantaneously, due to raindrop impact, in a mixing, or exchange, layer that sits just below the soil surface, and that there is no transport toward the mixing layer from deeper layers of soil [Ahuja, 1990; Ahuja and Lehman, 1983; Steenhuis et al., 1994a; Steenhuis and Walter, 1980; Zhang et al., 1999, 1999]. The diffusion approach suggests that solutes are transported from soil into runoff in a diffusion process, while ignoring the effect of raindrops [Wallach, 1991; Wallach et al., 1998; Wallach and van Genuchten, 1990]. Both approaches were successfully fitted to experimental data; however, assumptions made to ensure good fits either allowed for the theoretical mixing layer depth to exceed experimentally observed values or introduced immeasurable parameters. In essence, these efforts addressed two distinct mechanisms of solute transport with models that either explicitly allowed for only one, or incorporated multiple processes via inclusion of parameters with no clear physical definition [Ahuja, 1990; Steenhuis et al., 1994b; Steenhuis and Walter, 1980; Wallach and van Genuchten, 1990].

[79] From 2004 to 2007, Parlange and colleagues produced an influential body of work (currently at more than 80 ISI citations) that, for the first time, integrated raindrop-driven transport of solutes from the mixing layer into surface runoff, diffusion-driven transport from deeper soil layers into the mixing layer, and infiltration [Gao et al., 2004, 2005; Walter et al., 2007]. These processes were assumed to act in series and produced a superior fit to experimental data with no need for additional adjustable parameters.

[80] The conceptual schematic of model processes is shown in Figure 10. The soil-water system consists of three horizontal layers, namely the surface runoff (ponding water), the exchange layer, and the soil [Walter et al., 2007]. The exchange layer is assumed well mixed and serves as the intermediary compartment for vertical solute transport. Diffusion processes govern the transport of solutes from underlying soil into this layer where, in turn, raindrop impact governs the movement of solutes into runoff. Infiltration is also explicitly accounted for in this model.

[81] Governing equations were developed for each layer. Solute transport within the underlying soil is an advection-diffusion process driven by the upward movement due to diffusion and downward movement due to infiltration:

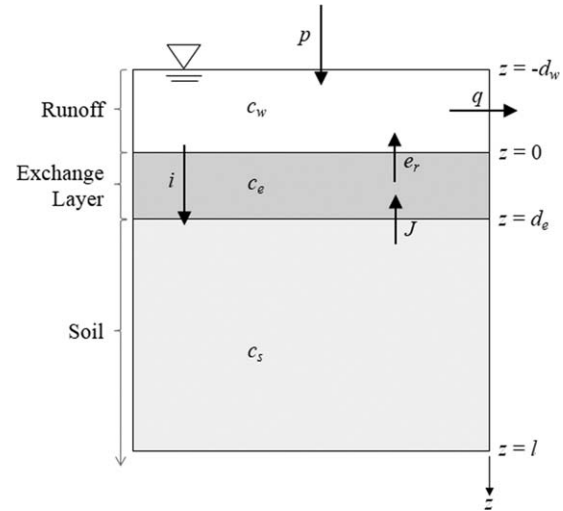


Figure 10. Conceptual schematic of the model's transport processes (after Gao et al. [2004] used with permission).

$$\frac{\partial(\theta_t c_s)}{\partial t} = \frac{\partial}{\partial z} \left(D \frac{\partial c_s}{\partial z} - I c_s \right), \quad (58)$$

where θ_t is the volumetric moisture content, and c_s is the chemical concentration in the soil-water below the exchange layer.

[82] Solute transport in the exchange layer includes diffusion from the soil layer below, raindrop-driven movement into runoff, and infiltration into the deeper soil:

$$\frac{\partial(\theta_t d_e c_e)}{\partial t} = F + (e_r + I)(c_w - c_e), \quad (59)$$

where d_e is the depth of the exchange layer, c_e is the solute concentration, c_w is the concentration in runoff water, e_r is the rate of soil water ejection into runoff due to raindrop impact, c_w is the concentration in water entering the exchange layer, and F is the diffusion rate of solute from underlying soil into the exchange layer, governed by Fick's law:

$$F = D \frac{\partial c_s}{\partial z}. \quad (60)$$

[83] Solute concentration in runoff is driven by raindrop impact from the exchange layer and by infiltration. Diffusion between the exchange layer and ponding/runoff water was neglected, giving:

$$\frac{\partial(d_w c_w)}{\partial t} + \frac{\partial(q c_w)}{\partial z} = e_r(c_e - c_w) - I c_w, \quad (61)$$

where d_w is the ponding water depth and q is the volumetric runoff flux per unit width. Overland flow was described by:

$$\frac{\partial d_w}{\partial t} + \frac{\partial q}{\partial x} = P - I. \quad (62)$$

[84] The model predictions were tested against experiments with [Ahuja and Lehman, 1983] and without [Gao et al., 2004, 2005] infiltration, in both cases with good agreement. The former demonstrated that infiltration reduced the

depth of the exchange layer, while the latter suggested that the assumption of a well-mixed exchange layer may not be realistic, especially in the early stages on rainfall when solute concentration in ejected soil water is near the initial condition value. It was also found that, after the initial flush, solute concentrations in runoff were controlled by diffusion of chemicals into the exchange layer and that the concentration in the exchange layer was different from that in runoff. These observations corroborated the dual mechanism of raindrop impact and diffusion governing chemical transport [Gao et al., 2004].

[85] The raindrop-driven exchange layer part of the model is conceptually akin to the HR soil erosion model in that it (1) reinterpreted rain-induced soil detachment as ejection of soil water from the soil during rainfall and (2) suggested that the mixing layer was equivalent to the “shield” produced when sediments detached from the soil surface deposited back and formed a protective layer that diminished the raindrop impact on the underlying soil surface [Hairsine and Rose, 1991; Sander et al., 1996]. Furthermore, the expression for e_r , the solute mass transfer rate due to raindrop impact, was developed based on a similar term for soil erosion [Hairsine and Rose, 1992b]:

$$e_r = \frac{aP}{\rho_b} \theta_i. \quad (63)$$

[86] The HR model was generalized in 1998 as a stochastic Markov model where soil particles alternated between rest and motion states [Lisle et al., 1998]. Interestingly, the same differential equations govern macroscopic variables, such as concentrations, and probability densities of individual soil particles. In this framework, averaging the stochastic motion of particles gives rise to deterministic HR model.

[87] Several aspects of the solute transport and erosion models were combined in the development of a multimedia stochastic model of microbial transport in surface flow, with application to *Cryptosporidium parvum* oocysts [Yeghiazarian et al., 2006]. This model was the first to consider microbial partitioning between solid and aqueous phases in surface water transport explicitly. Microbial transport was coupled with erosion because microorganisms are known to form bonds with soil sediments, and erosion often becomes an important vehicle of microbial mobility [Novotny and Olem, 1993]. Instead of two states of the stochastic erosion model (resting and moving), it employs five: microbes resting on the soil surface with and without attachment to soil particles, moving with surface flow with or without soil particles, and an absorbing state (sink in Figure 11) into which microorganisms transition via infiltration or biological decay (Figure 11). Transitions between these states are driven by dynamics of bond formation and breakup between soil particles and microbes, and, similarly to soil erosion, by microbial detachment from and deposition to the soil surface. The Markov process is described by

$$\frac{dp_{ij}(t)}{dt} + g_{ij}p_{ij}(t) = \sum_{l \neq j} g_{lj}p_{il}(t), \quad (64)$$

where g_{ij} , $i, j = 1, \dots, 5$ are transition rates of the process. Transition rates explicitly describe the physical and biological mechanisms that drive transitions between Markov states.

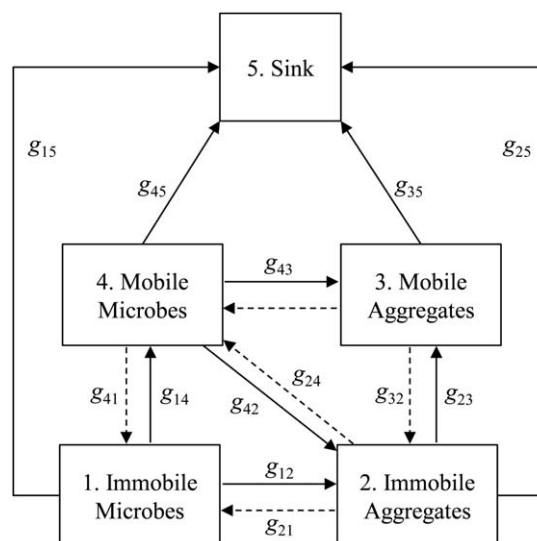


Figure 11. States and transitions of the Markov process that models microbe and sediment transport in overland flow, including exchange with the soil surface (after Yeghiazarian et al. [2006]).

For instance, g_{23} and g_{14} , the rates of mobilization of microorganisms from the soil surface, are functions of raindrop impact and the shear stress of the flow, and g_{i5} 's of biological decay. The model produced spatio-temporal distributions of probabilities of microorganisms being in different states. This information can be used for prediction of locations and time windows where the probability of finding microorganisms in runoff or on the soil surface is the highest.

[88] Non-point sources of contamination, in particular manure-fertilized agricultural fields, are primary contributors of microbial contamination in surface waters. In this light, modeling contaminant release from such sources is an important component in understanding overland microbial fate and transport and in developing better control solutions. One of the first works to look into this issue was Walter et al. [2001] who developed and experimentally tested a simple model of pollutant release from manure-like sources. In the model, the pollutant transport involves two independent processes: vertical advection-diffusion and/or dispersion in the downward direction, and horizontal advection at the bottom of the source (Figure 12). The source is assumed to be static and stable, with possibility of crust development.

[89] The horizontal advection at the bottom for the duration, t_b , until the bottom region is flushed of pollutants, is:

$$M_b = -cqrt, \quad (65)$$

where M_b is cumulative mass leaving from the bottom, w is the source width perpendicular to the flow, q is the discharge per width derived from St. Venant's continuity equation, t is time, and c is the concentration calculated from:

$$c = \frac{J_b}{P}, \quad (66)$$

where J_b is the rate of solute uptake from the source into the flow.

[90] When the source is not crusted, advection-dispersion dominates vertical transport from the upper part of the source to the bottom, the cumulative mass, M_{uc} , leaving the upper region is:

$$M_{uc} = cAPt, \quad (67)$$

where A is the constant horizontal cross-sectional area of the source, $c = c_0$ while pollutant is present in the upper region of the source, and $c = 0$ when it is not.

[91] When the source is crusted and there is no vertical flow of water through the source, diffusion dominates. The cumulative mass removed by diffusion is:

$$M_{ud} = 2Ac_0\sqrt{\frac{D}{\pi}(t - t_b)}. \quad (68)$$

[92] Experiments included laboratory studies with potassium chloride representing a conservative pollutant, and field studies with soluble reactive phosphorus at a manure-fertilized field in the Cannonsville watershed in the Catskills region (New York). Crusted, partially crusted, and uncrusted sources were used. All experimental data were in good agreement with model predictions. While further model improvements such as inclusion of a more sophisticated runoff component and of a dispersion mechanism were identified, the good data fit indicated that extending the model was not justified. An important conclusion of this study was that pollutant release from a fully crusted source is about 25–30% of that from an uncrusted source.

[93] The strength of this model lies in its simplicity and physical basis, allowing for further modifications to accommodate a wide range of test conditions and pollutants, should that be considered useful. While the theory is not limited to any specific species, the model was tested with conservative pollutants representing the worst-case scenarios. This choice was motivated by rising concerns about *Cryptosporidium parvum* oocysts, a waterborne microbial contaminant whose primary source is manure. The oocysts are resistant to harsh environmental conditions, nonreproductive outside their human and animal hosts and capable of causing large-scale outbreaks of gastrointestinal disease [MacKenzie et al., 1994; Walker et al., 1998].

[94] The presence of *Cryptosporidium* has been a major concern not only in surface water but also in ground water and soils [e.g., ten Veldhuis et al., 2010; Tufenkji and Elimelech, 2005; Tufenkji et al., 2004; Wilkes et al., 2009]. Rose et al. [1991] identified *Cryptosporidium* in well water and suggested a possible groundwater contamination route from sources on the soil surface, generating much interest in understanding oocyst transport in soil. Parlange and colleagues addressed this issue in a series of publications from 1999 to 2004 describing experiments and models of *Cryptosporidium* transport in saturated soils and in the vadose zone [Brush et al., 1999; Darnault et al., 2003, 2004]. These papers, currently at over 130 ISI citations, highlighted differences in microbial fate and transport in saturated versus unsaturated soils.

[95] A model of microbial transport in saturated soil columns was developed by Brush et al. [1999]. It employed a 1-D ADE [e.g., Parker and van Genuchten, 1984]:

$$R \frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial z^2} - v \frac{\partial C}{\partial z} - \mu C, \quad (69)$$

where C is the reduced concentration of liquid phase (dimensionless), μ is the first-order rate constant, and R is the retardation factor:

$$R = 1 + \frac{\rho_b k}{\theta_l}. \quad (70)$$

[96] Experiments were conducted to study *Cryptosporidium* transport in three fully saturated columns of glass beads, coarse sand, and shale aggregate. Experimental results fitted model predictions well ($R^2 > 0.87$). Fluctuations observed in effluent concentrations suggested that the ADE (69) may not have fully explained the transport mechanisms and that additional work was needed to better understand interactions between *Cryptosporidium* and various substrates. This conclusion was supported by differences in oocyst retardation among the three columns.

[97] The model of transport in the vadose zone assumed the presence of a distribution zone over the conveyance zone [Steenhuis et al., 1994a], whereby water and solutes move from the distribution zone through distinct paths of the preferential flow zone [Darnault et al., 2004]. Clearly, this approach has a similar mathematical flavor to that presented in section 3.3. The *Cryptosporidium* concentration in the distribution zone was modeled as:

$$C = C_0 \exp \left[-t \left(\frac{q}{W} + \beta \right) \right], \quad (71)$$

where C_0 is the initial concentration and β is the first-order removal rate from the solution. Removal due to adsorption to the air-solid-water interface was assumed irreversible and proportional to the concentration in solution. The concentration in the conveyance zone is then:

$$\frac{C(x, t)}{C_0} = \frac{1}{2} \exp \left[\frac{vx}{2D} (1 - \alpha') - t \left(\frac{q}{W} + \beta \right) \right] \operatorname{erfc} \left(\frac{x - vt\alpha'}{\sqrt{4Dt}} \right), \quad (72)$$

$$x + vt\alpha' > 3\sqrt{4Dt},$$

where

$$\alpha' = \sqrt{1 - \frac{4D(q + \beta W)}{v^2 W}}. \quad (73)$$

[98] The model output was compared to experimental results representing the worst-case scenario of *Cryptosporidium*-contaminated calf feces applied to the soil surface during rainfall. Transport with both fingered and macropore flow was explored. The model typically fit the *Cryptosporidium* breakthrough curves with $R^2 > 0.7$ and demonstrated the ability of pathogens to move in large numbers with preferential flow through unsaturated columns. The amount of oocysts in the effluent was much higher than the safe exposure limit.

[99] Comparison of experiments in saturated versus unsaturated conditions demonstrated that in flow through unsaturated columns the concentration in the effluent

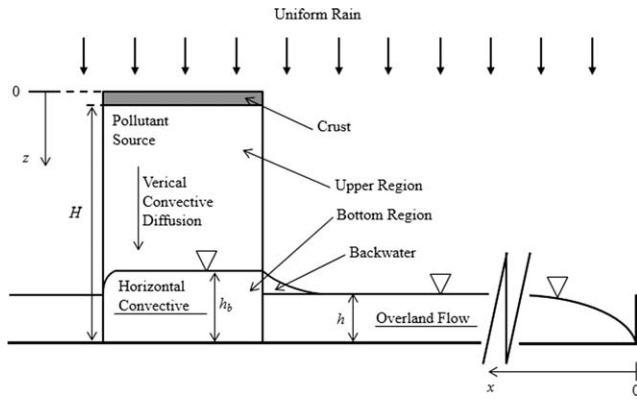


Figure 12. Conceptual model of pollutant release from manure-like source (after *Walter et al.* [2001] used with permission).

decreases rapidly, while in the saturated columns the breakthrough curves show a significant tail [*Brush et al.*, 1999; *Darnault et al.*, 2004]. These differences could be attributed to the differences in mechanisms of retention of *Cryptosporidium* in the soil, such as the presence of the air-water-solid interfaces in unsaturated columns that would not typically allow for remobilization versus the filtration mechanism in saturated columns that would likely allow remobilization.

6. Thermodynamics of Salt Solutions

[100] The blossoming of understanding of solute transport in porous media was largely driven by the need to predict contaminant transport, but Parlange’s personal interests were far more fundamental, seeing the connection between many hydrogeological processes mitigated by the complex solid-liquid-vapor interactions in porous media. In

many soil systems, salts can be found near saturation, or even in the form of pure crystals. These conditions are typically found where solid-form fertilizers are applied, or at evaporative surfaces of rocks and soils under saline conditions. Beyond the utility in predicting saline transport under these conditions, imposition of such a sharp transition in salt concentration could be used to elucidate vapor transport, ionic diffusion, and water transport in soils. The work of *Scotter and Raats* [1970] inspired Parlange to address this problem, seeking predictive (rather than descriptive) quantitative descriptions of these excellent data sets (which considered the movement of water and ions into a soil following an instantaneous imposition of a pure salt boundary condition). This analysis resulted in Parlange’s remarkable 1973 contribution that set the standard for simple and precise models for the evolution of salt, water, and vapor movement in the vicinity of soluble mineral contact with moist porous media [*Parlange*, 1973].

[101] *Scotter and Raats* [1970] plotted their data against the Boltzmann transform variable (η') of position (x , measured from the salt-soil interface) divided by the square root of elapsed time ($t^{1/2}$) to show that following an abrupt change in boundary condition (the addition of pure salt), water redistribution within the initially uniform semi-infinite column indeed followed the expected spatiotemporal structure of a diffusive system. The behavior of the system is revealed in Figures 13 and 14. In both the figures, the abscissa is the Boltzmann similarity variable based on data taken after 4, 5, 8, and 16 days in a set of meticulously conducted replicate experiments by *Scotter and Raats* [1970]. The data show that the interface between the initial and salty water is at $\eta' \approx 0.84 \text{ cm d}^{-1/2}$, which indicates the extent of the salt penetration.

[102] A key difficulty in modeling this system was that the magnitude of the response was greatest for intermediate

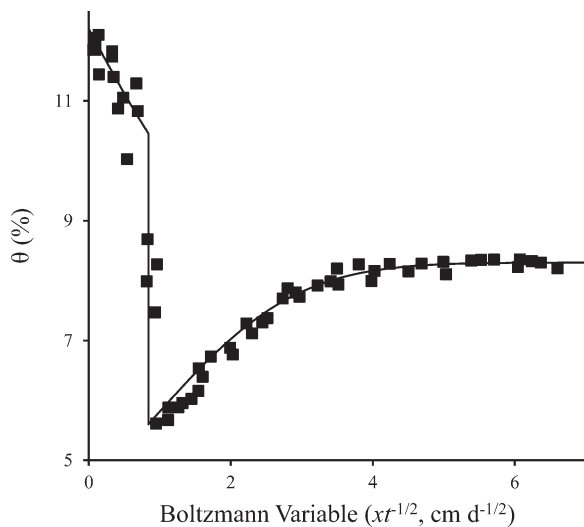


Figure 13. Gravimetric moisture content (line) from Parlange’s model of joint movement of salt and soil water following the application of pure salt at a boundary at $t=0$ (after *Parlange* [1973] used with permission), compared with the experimental data of *Scotter and Raats* [1970], shown as squares.

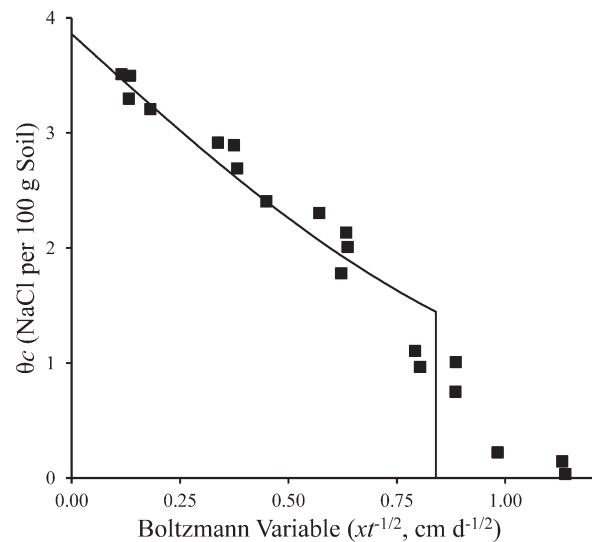


Figure 14. Salt mass as it varies with position (line) calculated from Parlange’s model of joint movement of salt and soil water following the application of pure salt at a boundary at $t=0$ (after *Parlange* [1973] used with permission). Solid squares show experimental data of *Scotter and Raats* [1970].

moisture contents, being smallest at either very dry or very wet conditions. Also, as just observed the problem contains a moving boundary dividing the region adjacent to the salt, where condensing water provides a layer of nearly saturated soil, and the source region, which is depleted of water through vapor loss to the saline vapor sink. As the process advances, the wetted region expands, and the boundary advances away from the salt. Parlange [1973] realized that the governing equations for water and salt movement must be solved for both regions and linked by the psychrometric equation and that high concentrations of salts should be expected to alter air-liquid interfacial energy. The seemingly intractable moving boundary problem yielded solution through insightful approximation. First, Parlange recognized that the deeply water-depleted soil from which vapor is drawn acts as a barrier to salt transport, so the salt distribution is only required for the near-salt region. Second, the water content of the near-salt region is nearly constant, with the salt source at one boundary, and the distribution of salt in the solution controlled by molecular diffusion. In the drying vapor-source area, as the largest pores are open, the gas diffusivity could be taken as constant with little loss of accuracy. The efficacy of these credible assumptions was borne out in the remarkable demonstration of the model provided by Parlange [1973], which compares well with the data of Scotter and Raats [1970], as shown in Figures 13 and 14. This work laid the foundation for the decades of exploration of these processes, which govern movement of solutes as a function of osmotic coefficient and water in deserts, near granular fertilizer, and at highly contaminated sites [e.g., Kelly *et al.* 1997; Kelly and Selker, 2001; Scotter, 1974; Weisbrod *et al.*, 2000]. It is striking that although these works add applications and influence of chemical characteristic of the salts in question, no significant advancements in the underlying assumptions or mathematical model have been required or developed since the 1973 paper.

[103] Typifying Parlange's influence on the field through collaboration, his invitation of then Ph.D. student Erik Burns to Cornell University for a one-month visit led to three publications that expanded the framework to a rigorous thermodynamic description of saline effects on constitutive soil-water relationships and permeability, illustrated for important salt solutions [Burns *et al.*, 2006a, 2006b, 2007]. This work continues to inspire efforts to model these complex systems, challenged to improve upon the precision achieved by Parlange four decades ago, exploring process descriptions and their physical basis through experiments of ever increasing sophistication [e.g., Gran *et al.*, 2011; Shokri *et al.*, 2009]. As is the case with so many of Parlange's contributions, in this area he advanced ideas that continue to intrigue and provide the basis for important lines of research in flow and transport in soils, setting a standard for excellence that keeps the work as relevant today as it was when first published.

7. Concluding Remarks

[104] Although we have only provided a brief overview of Parlange's contributions in the areas of solute and sediment transport, it is evident that their scope is both broad and deep. Parlange's emphasis is on combining physical understanding with theoretical modeling. The results are clarifica-

tion of mechanisms and mathematical results that are both insightful and of practical use. He has demonstrated many times that this *modus operandi* is highly beneficial in uncovering insights that lead to scientific advancement.

Notation

a	Uneroded soil detachability parameter (function of h), ML^{-3}
a_0	Threshold value of a indicating when erosion occurs in uneroded soil, ML^{-3}
a_d	Detachability parameter of the deposited layer (function of h), ML^{-3}
a_{d0}	Threshold value of a_d indicating when erosion occurs in previously eroded soil, ML^{-3}
A	Constant horizontal cross-sectional area of the source, L^2
ADE	Advection-dispersion equation
A_w	Proportion of soil that is wet
$A_{w,max}$	Maximum proportion of soil that is wet
B	Infiltration rate or evaporation rate, LT^{-1}
c	Solute concentration, ML^{-3}
c_0	Influent concentration, ML^{-3}
c_e	Solute concentration in the exchange layer, ML^{-3}
c_f	Flux concentration, ML^{-3}
c^{sp}	Total suspended sediment concentration, ML^{-3}
c_i^{sp}	Suspended sediment concentration for size class i , ML^{-3}
c_i^{sp-}	See (48), ML^{-3}
c_i^{sp+}	See (49), ML^{-3}
c_r	Resident concentration, ML^{-3}
c_s	Chemical concentration in the soil water below the exchange layer, ML^{-3}
c_w	Solute concentration in runoff water, ML^{-3}
C	Reduced concentration in the liquid phase
C_0	Initial (dimensionless) concentration
d	Depth of the distribution zone, L
d_e	Depth of the exchange layer, L
d_i	Deposition rate of size class i , $\text{ML}^{-2}\text{T}^{-1}$
d_w	Ponding depth, L
D	Diffusion/dispersion coefficient, L^2T^{-1}
D_{ap}	Apparent diffusion/dispersion coefficient, L^2T^{-1}
D_R	Rooting depth, L
D_I	Raindrop detachment rate, $\text{ML}^{-2}\text{T}^{-1}$
D_F	Flow detachment rate, $\text{ML}^{-2}\text{T}^{-1}$
e_{di}	Rainfall detachment rate of the deposited layer of size class i , $\text{ML}^{-2}\text{T}^{-1}$
e_i	Rainfall detachment rate of the uneroded soil of size class i , $\text{ML}^{-2}\text{T}^{-1}$
e_r	Rate of soil water ejection into runoff due to raindrop impact, $\text{ML}^{-2}\text{T}^{-1}$
E	Evapotranspiration rate due to plants, LT^{-1}
f	Dimensionless function (11)
F	Diffusion rate of solute from underlying soil into the exchange layer, $\text{ML}^{-2}\text{T}^{-1}$
F_r	Fraction of excess stream power effective in entrainment
g	Magnitude of gravitational acceleration, LT^{-2}
g_{ij}	Transition rate in a Markov process, T^{-1}
G	Erosion source or sink term, $\text{ML}^{-2}\text{T}^{-1}$
h	Overland flow depth, L
\bar{h}	Threshold depth of the detachment rate, L

h_i	Water depth where characteristic branches meet, L	z	Soil depth, L
h_0	Water depth at $x = 0$, L	z_f	Front position, L
H	Protection factor provided by the deposited layer	1-D	One-dimensional
H'	See (50)	α	See (26)
H^+	See (41)	α'	See (73)
HR	Hairsine-Rose	β	First-order removal rate from the solution, T^{-1}
i	Specifies a given sediment size class	Δx	Increment in the x direction, L
I	Net water flux at the soil surface, LT^{-1}	Δz^*	Movement of water front position due to water input at the soil surface, L
J	Water flux, LT^{-1}	χ	Dispersivity, L
J_b	Rate of solute uptake from the source into the flow, $ML^{-2}T^{-1}$	δ	Soil characteristic parameter
J_e	Specific energy of entrainment, M^2T^{-2}	η	a_d/m^* , L^{-1}
k	Adsorption partition coefficient, L^3M^{-1}	η	Boltzmann variable, $L^{-1/2}$
K_s	Saturated hydraulic conductivity, LT^{-1}	θ	Average volumetric moisture content in the root zone
L	Length of the flow domain, L	θ_d	Volumetric moisture content in the distribution zone
L_b	Wetted bulb length, L	θ_f	Moisture content in a finger
L_s	Characteristic length scale over which a liquid sample is withdrawn in a porous medium, L	θ_{fc}	Field capacity
m	Total deposited mass per unit area, ML^{-2}	θ_J	θ_{fc} or θ
m_i	Deposited mass per unit area of sediment in size class i , ML^{-2}	θ_s	Saturated volumetric moisture content
m_{i0}	Initial value of m_i , ML^{-2}	θ_t	Volumetric moisture content
m^*	Mass per unit area of the deposited sediment required to protect the original soil, ML^{-2}	λ	Interpolation parameter
m^{sp}	Mass per unit area of suspended sediment (57), ML^{-2}	μ	First-order rate constant, T^{-1}
M	Mass of chemical per unit area, ML^{-2}	ν_i	Fall velocity for sediment size class i , LT^{-1}
M_b	Cumulative mass leaving from the bottom of the sink, M	ρ	Water density, ML^{-3}
M_{uc}	Cumulative mass leaving the upper region, M	ρ_b	Soil bulk density, ML^{-3}
M_{ud}	Cumulative mass removed by diffusion, M	ρ_s	Particle solid density, ML^{-3}
n	Manning's roughness coefficient, $TL^{-1/3}$	ψ	Matric potential in a finger, L
N	Number of sediment particle sizes	ω_i	See (47)
p_i	Proportion of sediment in size class i	Ω	Stream power, MT^{-3}
p_{ij}	Transition probability function in a Markov process	Ω_{cr}	Critical stream power, MT^{-3}
P	Rainfall rate, LT^{-1}		
Pe	Péclet number		
Pe_{ap}	Apparent Péclet number		
q	Discharge rate per unit width, L^2T^{-1}		
q_s	Sediment flux, $ML^{-1}T^{-1}$		
r_i	Sediment entrainment rate (flow-driven detachment) from the uneroded soil for sediment size class i , $ML^{-2}T^{-1}$		
r_{di}	Sediment entrainment rate (flow-driven detachment) from the previously eroded soil for sediment size class i , $ML^{-2}T^{-1}$		
R	Retardation factor		
s	$hc_i^{sp} + m_i$		
S_0	Bed slope		
t	Time, T		
t_b	Flushing time, T		
t_s	Sampling time, T		
t_0	Arbitrary initial time, T		
T_c	Transport capacity, $ML^{-1}T^{-1}$		
v	Advection velocity, LT^{-1}		
ν_i	Fall velocity for size class i , LT^{-1}		
V	Finger speed, LT^{-1}		
w	Source width perpendicular to the flow, L		
W	Apparent water depth in the distribution zone, L		
x	Horizontal or downslope distance, L		

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