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Spatial Variability of Solute Transport Mechanisms Based on Generalized Transfer Function Model

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Abstract. The flexible generalized transfer function model (GTF) and TDR based time normalized resident concentrations were combined in order to characterize solute transport mechanism both at local and field scale. A leaching experiment was carried out in a plot under greenhouse, where TDR probes were installed at three different depths at 37 sites along a 40 m transect. The field plot was brought to steady-state water content; a pulse application of 3.87 mm of KCl solution was applied. Measurements of water content (θ) and impedance (Z) were simultaneously taken to follow the KCl solution propagation through the soil profile. Time series of relative resident concentrations for each site where effectively interpreted in terms of GTF model. The field scale behavior was described by calculating a local average and an integral average, by averaging local scale parameters and local scale original measurements, respectively. The two different averaging schemes resulted in two significantly different field scale solute transport behaviors.

Keywords. Soil water content – Solute transport models – Time Domain Reflectometry (TDR) – Spatial variability.

Variabilité spatiale des mécanismes de transport des solutés basée sur la Réflectométrie dans le Domaine Temporel et la Fonction de Transfert Généralisée

Résumé. *Ce travail associe la fonction de transfert généralisée flexible (modèle GTF) et les concentrations massiques normalisées à l'égard du temps se basant sur la Réfléctométrie dans le Domaine Temporel (TDR), pour caractériser le transfert des solutés dans le sol. Un essai de lessivage a été mené sur sol dans une parcelle de 40 m de long sous serre où des sondes TDR ont été installées à trois différentes profondeurs à 37 points le long de la parcelle. La teneur en eau du sol a été ramenée à l'état d'écoulement permanent; Une impulsion de 3.87 mm de solution de KCl a été appliquée. Les mesures de la teneur en eau (θ) et d'impédance (Z) ont été prises simultanément pour suivre la propagation de la solution de KCl le long du profil du sol. Les concentrations massiques relatives à chaque point ont été caractérisées avec succès par le modèle GTF. Les moyennes des paramètres locaux (moyenne locale) et des mesures locales (moyenne intégrale) ont été estimées pour représenter le transfert des solutés et les paramètres correspondants a l'échelle parcellaire. Les deux moyennes ont montré deux comportements de transfert des solutés à l'échelle parcellaire très différents.*

Mots-clés. *Teneur en eau du sol – Modèles de transfert des solutés – Réflectométrie dans le domaine Temporel (TDR) – Variabilité spatiale.*

I – Introduction

In the last decades, particular attention has been focused on the hazards posed by widespread pollution of groundwater resources, which are especially vulnerable due to the extent of the surface area directly affected by land use (Jury *et al.*, 1991; Carravetta, 1996). Major progresses have been achieved through parallel studies in setting up experimental techniques for monitoring inorganic, organic and biological substances to be found in soils and aquifers (Kachanosky *et al.*, 1992; Comegna *et al.*, 1999), using mathematical models and having widespread recourse to numerical calculations (Santini, 1992), as well as characterizing natural systems on a statistical basis (Jury *et al.*, 1987; Kutilek and Nielsen, 1994; Comegna and Vitale, 1993). However, according

to most recent studies, it is becoming increasingly clear that such models fail to exhaustively describe the phenomena in their totality and are also difficult to apply on a regional level because of the heterogeneity of natural porous media and the large number of chemical, physical and biological parameters to be considered (Jury and Flühler, 1992). The heterogeneity of natural porous media may limit the applicability of the CD equation in the field. In such conditions, an alternative path to describe the transport of solutes is the stochastic–convective approach, SC, in which the solute is assumed to move in isolated stream tubes at different velocities without any lateral mixing (Dagan and Bresler, 1979). The effectiveness of this approach in the field has been amply demonstrated (Jury, 1982; Butters and Jury, 1989; Heuvelman and McInnes, 1999). The SC approach was also applied successfully at a laboratory scale on undisturbed soil columns (Khan and Jury, 1990; Comegna *et al.*, 2001). The complex heterogeneity of soil has encouraged the development of transport theories based on conceptual models, such as the transfer function approach (Jury, 1982). A transfer function model can predict the flux density from a system with definite boundaries depending on the input flux without any need to describe the complex process that takes place within the porous system. Jury (1982) suggested a log-normal travel time probability density function and set up the convective log-normal transfer function model (CLT). This model was shown to supply an accurate prediction of solute transport in field experiments. The CDE and the stochastic–convective model represent two extreme processes for solute dispersion. In the CDE, perfect lateral mixing is assumed, whereas the stochastic–convective process assumes that the solute moves at different velocities in isolated stream tubes without lateral mixing. These models are limited to describing solute transport characterized by either a linear or a quadratic increase in the travel time variance with depth (Liu and Dane, 1996). However, solute transport in heterogeneous porous media cannot always be conceptualized as being either a convective–dispersive or a stochastic–convective process. Therefore, it is necessary to develop a general model that can describe not only the two extremes, but also other transport processes. Zhang (2000) presented a flexible generalized transfer function models (GTF). The GTF is a four-parameter flexible transfer function able to describe both the convection-dispersive (CD) and the stochastic-convective (SC) process of dispersion in a soil. In additional the GTF model can also be used to characterize solute transport processes in heterogeneous soils such as those in which the mean travel time increase non-linearly with depth and those in which the dispersivity is a scale dependent function. The GTF is thereby a comprehensive transfer function model allowing the solute transport process in heterogeneous soils to be formalized in a synoptic way. Regarding solute transport modeling several researches have calculated values of local and field-scale dispersion behavior during unsaturated flow. In a stony soil field-scale dispersivities were four times greater than the average local-scale dispersivity (Schulin *et al.*, 1987). Similar findings were reported by Butters et al. (1989), who observed that the field scale variance of the field average solute BTC was twice as great as the average local-scale variance.

To date, no method has been proposed for determining the scale at which the true field-scale variance is reached. For monitoring and modeling at the field scale, it is important to know the scale dependence of solute travel-time variance to accurately characterize transport phenomena at this site. Given the expected variation in the soil properties in space and time (Mulla and McBratney, 1999), we must develop effective mean to characterize them across representative field areas.

Recently, great progress has been made in identifying transport processes by applying TDR technology to measure solute concentration (see, e.g., Vanclooster *et al.*, 1995; Coppola and Damiani, 1997; Comegna *et al.*, 1999). Major advantages of using TDR technology for characterizing solute are related to the possibility of easily applying it to undisturbed soil material and of measuring transport with a high spatiotemporal resolution. This latter property is a prerequisite for validating transport concepts for undisturbed soil. A major problem in the earlier studies of solute transport with TDR was the need for a soil and layer specific calibration equation, relating signal attenuation to the resident solute concentration of an ionic tracer (Mallants *et al.*,

1996; Vanclooster *et al.*, 1994; Vogeler *et al.*, 1997). However, to avoid this, Vanderborght *et al.*, (1996) introduced the time-normalized resident concentration in terms of the two-parameter CLT solute transport model, which allows the CLT parameters to be directly identified from TDR output readings. Jacques *et al.*, (1998) extended the approach for the two-parameter CD model.

Finally, Javaux and Vanclooster (2003) presented an analytical solution for time normalized resident concentration for the GTF model. A unique advantage of TDR is its ability to rapidly measure both resident concentration of a solute and water content with the same probe and in the same sampling volume.

The general objective of this study is to improve our understanding about the link between the small scale variability of the local transport properties and large scale transport behavior. The structure of solute transport mechanism variability in the soil will be drawn up by integrating TDR based resident concentrations measurements and the Generalized Transfer Function model.

Firstly, the local solute transport mechanism will be characterized on several sites along transect area by using TDR probes at different depths and in terms of GTF model.

Second, The local scale measurement and parameters will be integrated along the whole transect and normalized for the different local water contents, in order to build a time-integral normalized resident concentration at each depth to be used as field-scale curves.

II – Materials and methods

1. Experimental procedure

A snapshot of the experiment layout is given in figure 1. The experimental layout consists of irrigation system, TDR probes and the drainage system. The experimental area was covered by plastic mulch in order to avoid soil evaporation and to assure only downward movement of the applied water and solution. Then the drip irrigation was adapted to apply the rate of 10 mm/day at 5 a.m.. An automatic irrigation scheduler was used to maintain the desired rate.

Figure 1. The experimental layout.

The field plot was pre-irrigated with fresh water having an EC of 1.05 dS/m until a steady state water condition was attained.

After reaching the steady state condition, a depth of 3.87 mm of KCl solution (23.5 g/l with specific mass of 50 g/m2 of Cl-) was applied through the drip irrigation system as a pulse application (δ-Dirac type top boundary condition), while keeping the same supply rate of 10 mm/day.

Then, the fresh water was newly applied to clean the drip system and to force the KCl solution downward into the soil.

The volumetric water content, θ, and the impedance, Z, monitored before applying KCl solution were considered as initial values for the experiment.

After KCl solution application, daily measurement of θ and Z were taken in the morning and evening to follow the KCl solution propagation through the soil profile.

A time domain reflectometry (TDR100) device and TDR probes were used to measure the water content and the impedance.

The transmission line consisted of an antenna cable (RG58, 50Ω characteristic impedance, 210 cm length and with 0.2Ω connector impedance) and of three wire probe, 15 cm length, 2 cm internal distance, 0.3 cm diameter. The TDR probes were inserted at three different depths in 37 sites, at 1 m intervals, along the transect. The surface probes were installed vertically (0-15 cm) while the deeper probes were inserted horizontally at 20 and 40 cm from the soil surface.

III – Results and discussion

1. Actual situation of the reservoir R.1

The experiment was realized under steady-state water contents in the soil profile compatible with the characteristics of the different sites and with the boundary conditions adopted.

For each position where TDR probes were located, a temporal series of water content were obtained as shown as Figure 2 (a, b, c) which describes the water content variability along the transect at the three different times, the initial time, the final time, the middle time.

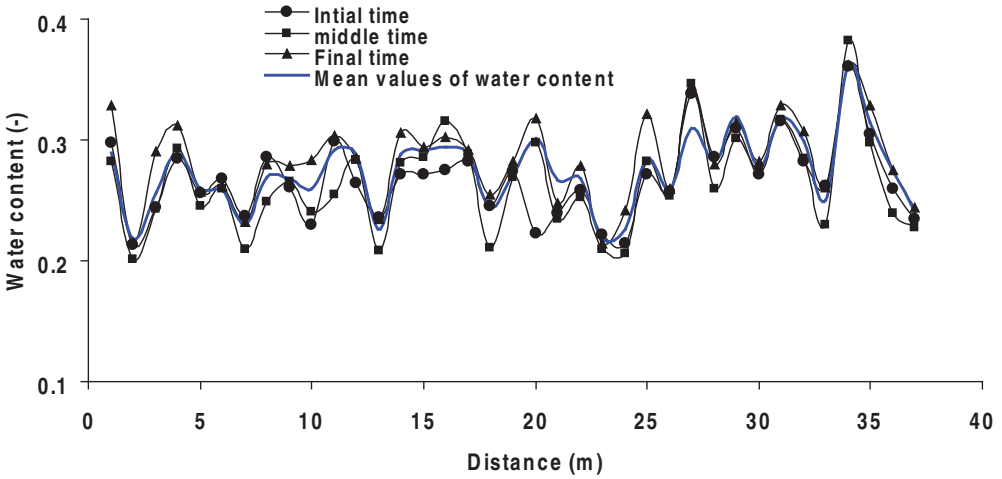

Figure 2a. Water content variability along the transect at 0-15 cm observation depth, at three different times, the initial time, the final time, the middle time.

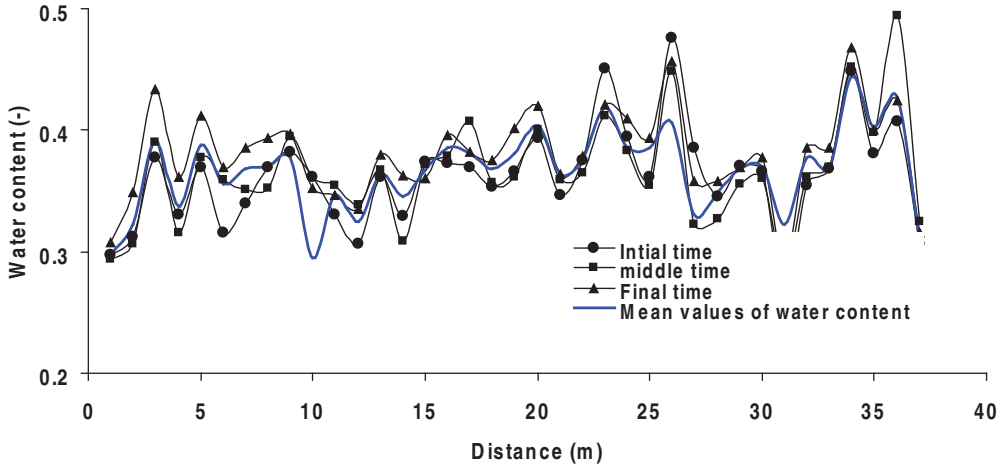

Figure 2b. Water content variability along the transect at 20 cm observation depth, at three different times, the initial time, the final time, the middle time.

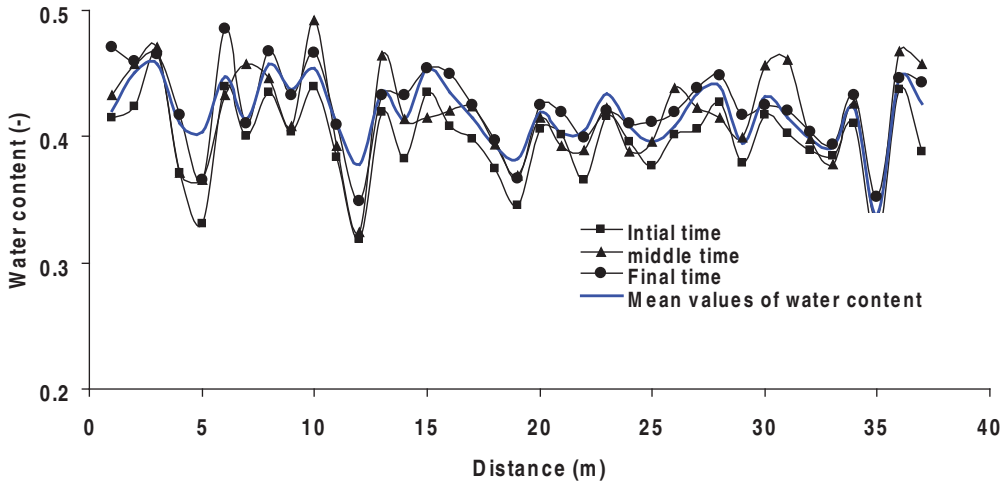

Figure 2c. Water content variability along the transect at 40 cm observation depth, at three different times, the initial time, the final time, the middle time.

The mean values of water content are also shown. Notwithstanding the variability along the transect, which is strictly related to the variability of soil physical characteristics and to some differences in the supply rate along the drip lines, what should be noted is the relatively constant water content for each location. This is confirmed for each depth. This result suggests that the steady-state conditions required for the experiment were effectively fulfilled, but it also implies that the important variability of the water content along the transect can not be neglected and has to be explicitly taken into account in all the calculations require use of the measured water contents.

The solute propagation along the soil profile was monitored by measuring TDR impedance loads at large times. Just as an example, the temperature corrected TDR impedance loads as a function of time for the 25cm and 40 cm depths of the measurement site labelled as site 18 are shown in Figure 3.

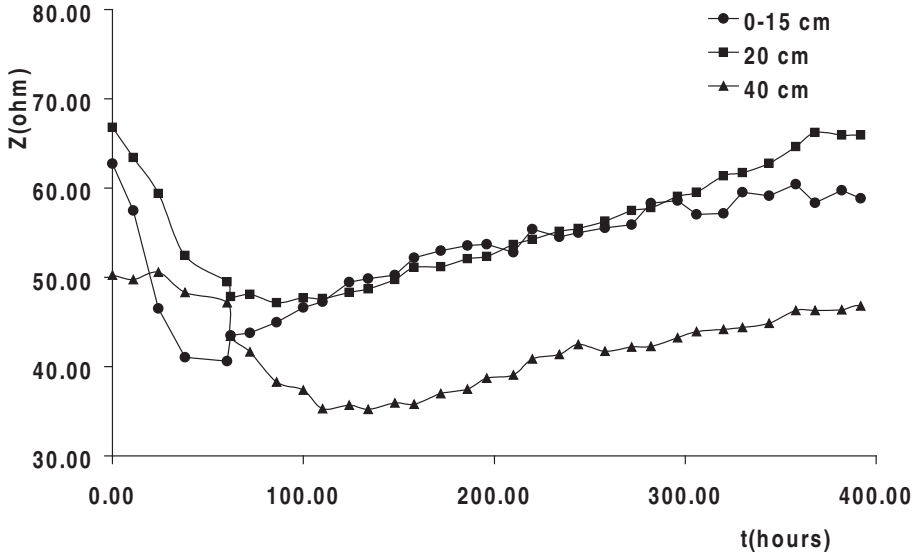

Figure 3. Temperature corrected TDR impedance loads for the three depths of site 18.

The impedance load changed more gradually while the solute reached in sequence the other two measurement depths. A different propagation velocity was observed for different locations. Such differences may be attributed to the different physical characteristics of the soils examined.

The reflectograms were than used for calculating time normalized resident concentrations, Crt*, for each depth and for each site according to the method given by Vanderborght et al. (1996). The pertaining curves for the site 18 are shown in Figure 4.

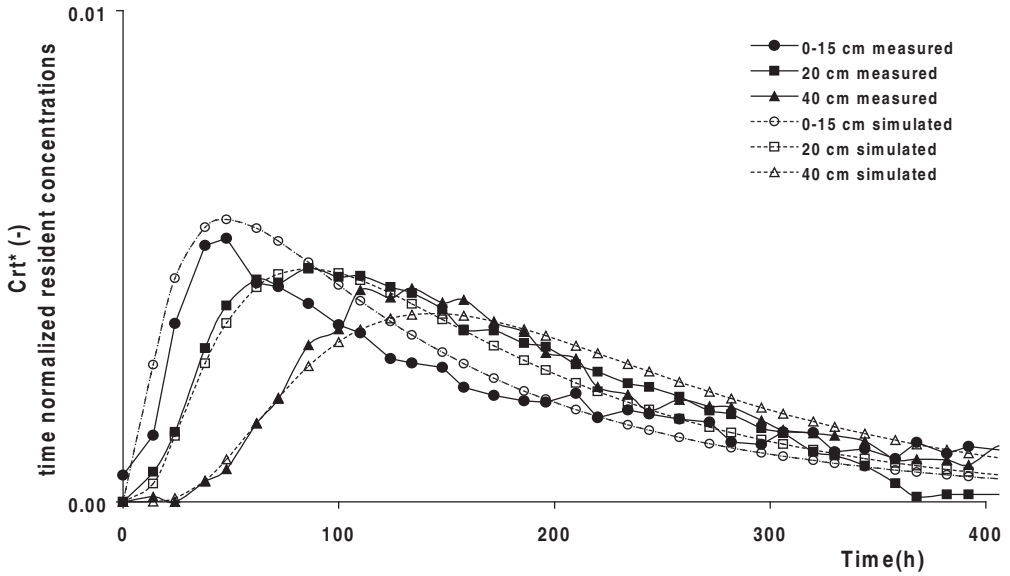

Figure 4. Observed and fitted normalized resident concentrations, Crt*, for site 18.

GTF parameters were obtained by fitting the following nonlinear four-parameter function:

$$C^{rt*}(z,t) = \frac{a\ln(t) - a\sigma_z^2 - a\mu_z + \lambda_1}{t\lambda_1\sigma_z\sqrt{2\pi}} \exp\left[-\frac{(\ln(t) - \sigma_z^2 - \mu_z)^2}{2\sigma_z^2}\right]$$
$$a = \frac{(\lambda_2 - \lambda_1)}{\sigma_z^2}\left[1 - \frac{1}{\exp(\sigma_z^2)}\right] \quad (1)$$

to the Crt* curves (see Figure 4). In equation 1, μ and σ are the mean and standard deviation of lnt and λı and λ2 are parameters of the time moments (Zhang, 2000).

In order to identify robustly the governing transport mechanisms, the least square optimization simultaneously were applied to all the three Crt* curves measured at three different depths. Since the model is able to describe a wide range of dispersion processes evolving with depth, fitting the model to all the depths together resulted in only one parameter set through which the transport process could be directly identified. The difference between the two fitted parameters, λ1 and λ2, characterizes the evolution of the dispersivity with depth while λ1 provides information on the variation of solute velocity with depth.

Thus, transport mechanisms can be classified in function of the difference λ1- λ2: i) if λ1-λ2=0, that is the variance of the ln(t) is constant with travel distance, the process is SC; ii) if λ1-λ2<0, the variance of ln(t) increases with travel distance and the process is scale dependent; iii) if λ1-λ2>0, the variance of ln(t) decreases with travel distance as in the CD model (λ1-λ2=0.5).

In the same way, transport processes can be classified in terms of the relationship between dispersivity, α, and depth. In this study the dispersivity was calculated in terms of GTF parameters. The convection-dispersion (CD) and the stochastic-convective with log-normal distribution of travel times (SC) models represent two radically different solute transport mechanisms. The models respectively conceptualize independent and correlated solute travel times with depth. The depth dependency of the dispersivity will reveal whether the solute transport process meets the CD or the SC assumptions. A linear increase of α(z) for SC and constant α(z) for the CD are expected. In some cases the variance of ln(t) increases with the travel distance and a scale-dependent transport process different from CD and SC should be used.

Thus, a plot of α(z) enables us to identify whether the solute transport mechanism meets the CD or the SC assumptions or whether different scaling parameters for the travel time distribution moments should be used.

Figure 5 shows the evolution with the distance of the λ1-λ2 values obtained along the transect.

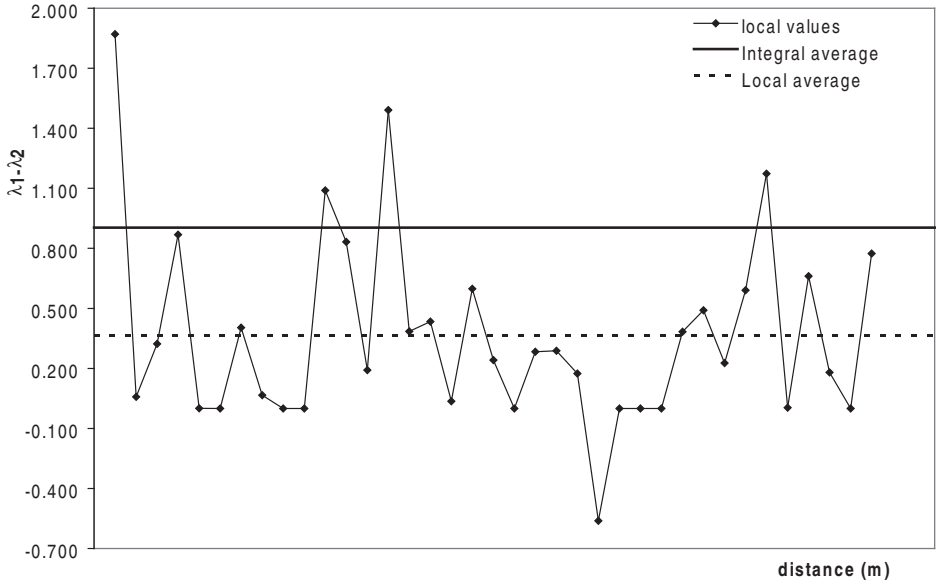

Figure 5. The evolution with the distance of the λ1-λ2 values obtained along the transect.

The maximum value observed was of 1.87 and the average was 0.366.

The difference is at all but one site positive, thus indicating a transport mechanism which is in general of the CD type. Nevertheless, if one couples the graph in Figure 5 and graph in Figure 6, showing the dispersivity as a function of the depth for three sites along the transect one can see that the transport process is frequently a scale dependent process with a contrasting behaviour. In some cases the dispersivity is constant with depth, as in the CD model, while in other cases dispersivity may increase (as predicted by the SC model) or even decrease with depth.

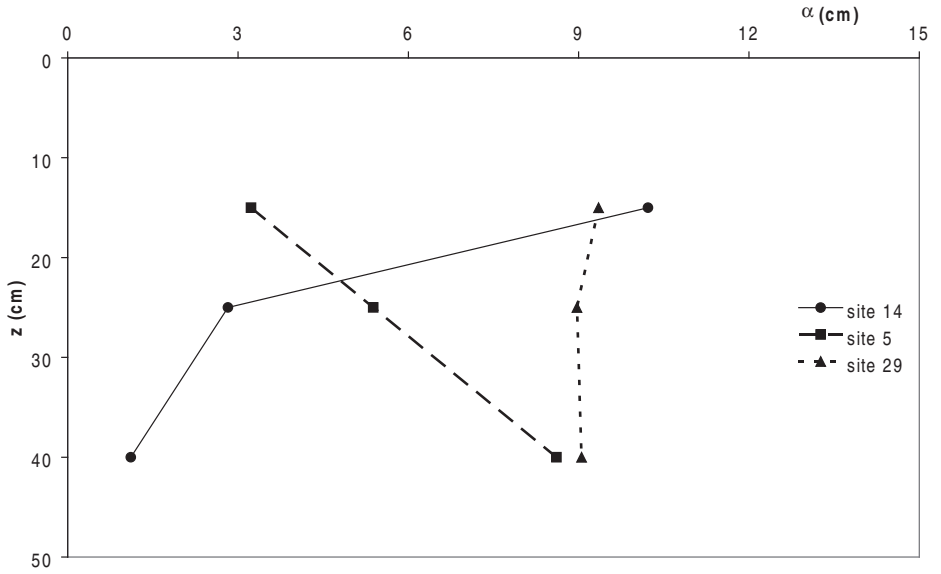

Figure 6. The dispersivity as a function of the depth for three sites.

Figure 7 shows the evolution with distance along the transect of the dispersivity at the three depths.

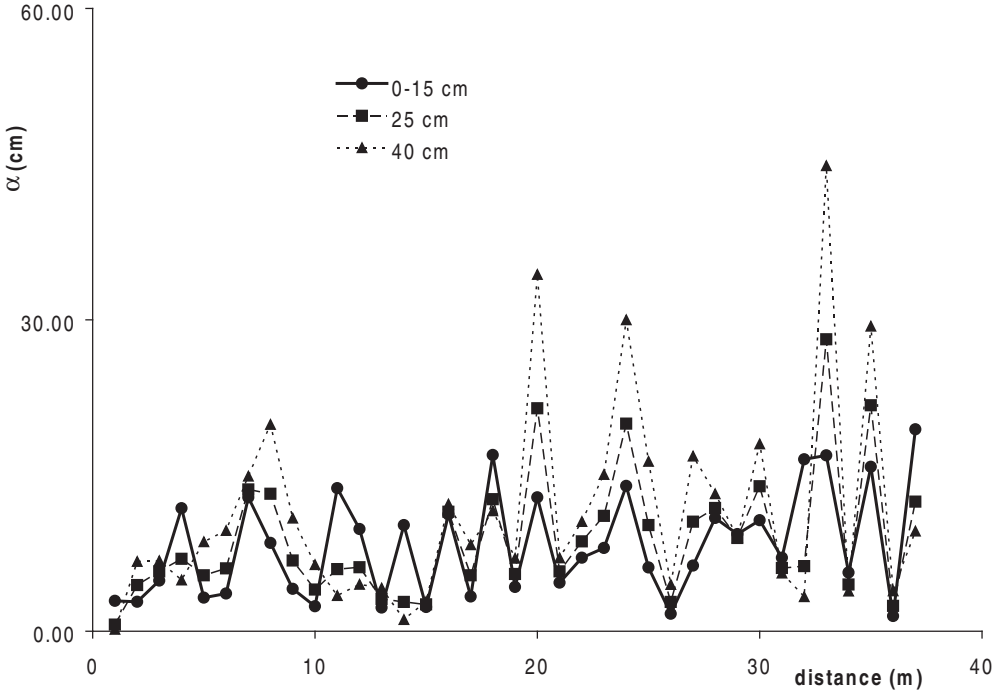

Figure 7. The evolution of dispersivity along the transect at the three observation depths.

The figure shows generally increasing values of dispersivity with depth (SC transport mechanism). However, there are cases were an inverse behaviour was observed. The values of dispersivity at 40cm allow also to discriminate two different zones along the transect, one with relatively small values and one with large values of dispersivity. As dispersivity is generally considered an intrinsic property of porous media related to the soil particle aggregation and the consequent pore-size distribution, higher values of the parameter could be related to a better soil structure, even at large depths, in the second part of the transect. Like for the water content, variograms for the dispersivity showed no significant spatial dependence for all depths, beyond the minimum separation distance of the TDR probes (1m).

However, one should be aware that estimating soil solute transport parameters at local scale is only a first step toward the prediction of solute transport at field scale, which is the shortest scale of applicative interest. Extrapolating local scale solute transport experiments to field situations remain a complex issue because of the natural heterogeneity of soils. Accordingly, the second objective of this work was to assess the transition from the local scale to the field scale during unsaturated flow conditions.

Two limiting horizontal spatial scales were considered in terms of solute transport mechanism and parameters: the local scale and the field scale. The field scale behavior was analyzed in terms of what we call a local parameter average and an integral measurement average.

The first one was obtained by determining the local scale solute transport mechanism and related parameters for each location and then by averaging the parameters over locations. The average value (0.366) for $\lambda 1$-$\lambda 2$ is shown as a dashed line in figure 5.

Figure 8 shows the local average dispersivity values in function of depth, z (8.28, 8.91 and 11.35 cm for the depths of 0-15, 25 and 40 cm, respectively).

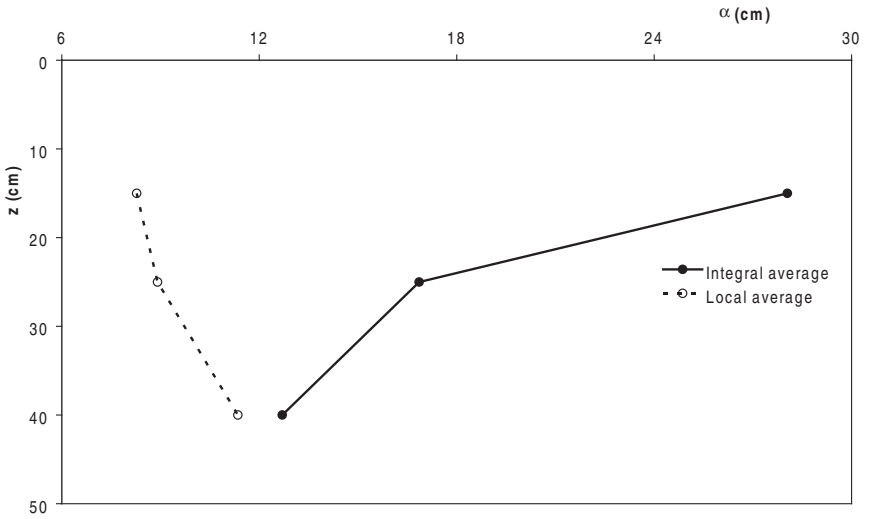

Figure 8. Local and integral average dispersivity values as a function of depth.

It can be seen that the dispersivity behaviour follows a SC transport mechanism even if it seems to start as a CD like mechanism. A dispersivity increasing with depth was observed.

The integral measurement average was obtained from the same measurements by firstly averaging the time normalized resident concentrations to get a single global curve for the whole transect and then evaluating the global scale solute transport mechanism and related parameters.

Figure 9 shows the integral average curves for the three depths, along with the best fitting obtained in terms of GTF parameters.

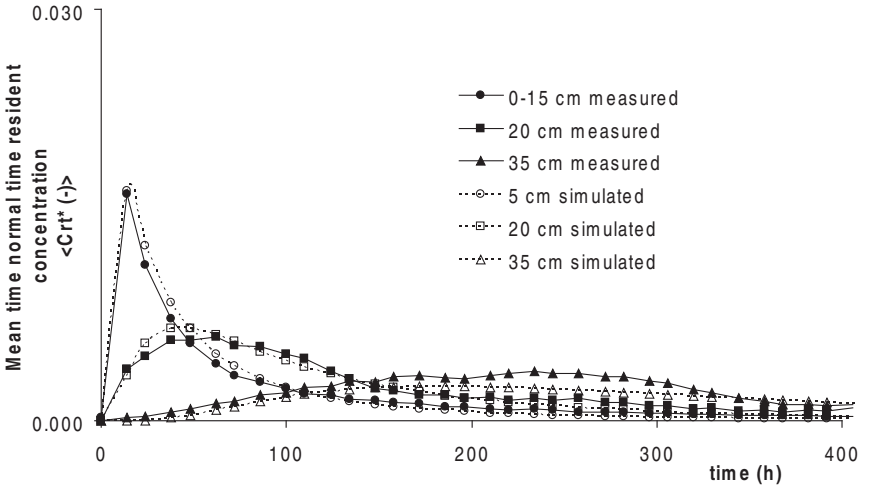

Figure 9. The integral average curves for the three depths, along with the best fitting obtained in terms of GTF parameters.

A difference $\lambda 1$-$\lambda 2$=0.904 was obtained, along with integral average values of dispersivity of 28.05, 16.86 and 12.70 cm, for the three depths, respectively. These dispersivity values are plotted in figure 8.

In general, the comparison between local average and integral average approach results in a global process (as indicated by the corresponding $\lambda 1$-$\lambda 2$ values, which differ significantly). Concerning the dispersivity, a contradictory behaviour has been estimated with the two different averaging approaches. The integral average approach results in dispersivity values decreasing with depth, thus resulting also in very different predictions of solute travel time distributions.

The only way for solving the complex dilemma of choosing the appropriate averaging scheme the local scale behavior (measurement or parameters) for a correct transition from the local scale to the field scale behavior would be a comparison of measurement at different scales.

Unfortunately, this study was not a true comparison of measurements at different scales but rather a comparison of estimates of parameters at two scales based on measurements at single locations. Anyway, the results confirm that the transfer of information (upscaling) from the local scale to a larger scale is a practice which remains to a large extent unresolved due mainly to the complex variability of soil hydrological properties. Understanding this transition is crucial to effectively predict transport processes for applicative uses.

IV – Conclusion

TDR measurements at the local scale were used to estimate solute transport parameters at both the local and field scale. In a sense, this study was not a true comparison of measurements at different scales but rather a comparison of estimates of parameters at two scales based on measurements at single locations.

In this study, closed-form expressions for the time normalized resident concentrations, Crt*, for the generalized transfer model (GTF model) have been applied. The Crt* pdf was fitted to the measurements in all the 37 sites and gave excellent results. This means that with real data, the transport process can be directly identified by observing the $\lambda 1$and $\lambda 2$ fitted parameters. The difference between these two parameters characterizes the evolution of the dispersivity with depth while $\lambda 1$ provides information on the variation of solute velocity with depth.

The least square optimization was applied simultaneously to all the three Crt curves measured at three different depths. Since the model is able to describe a wide range of dispersion processes evolving with depth, fitting the model to all the depths together resulted in only one parameter set which should have a better predictive power.

The additional parameters $\lambda 1$ and $\lambda 2$ can be robustly estimated as the information on the propagation mechanism is significantly carried by the three curves used simultaneously in the fitting procedure.

With the local scale parameters at hand, there is the successive problem of defining an aggregation rule to obtain large scale parameters for large scale simulations. This frequently involves a transfer of information (upscaling) from the microscale to a larger scale, a practice which remains to a large extent unresolved due mainly to the complex variability of soil hydrological properties.

Two limiting horizontal spatial scales were considered in terms of solute transport mechanism and parameters: the local scale and the field scale. The field scale behavior was analyzed in terms of what we called a local parameter average and an integral measurement average. The first one was obtained by determining the local scale solute transport mechanism and related parameters for each location and then by averaging the parameters over locations. The second one was obtained from the same measurements by firstly averaging (taking into account the different water content measured at each location) the time normalized resident concentrations to get a

single global curve for the whole transect and then evaluating the global scale solute transport mechanism and related parameters. It was shown that the average of the local set of parameters does not represent effectively the integral average of the measurements.

The only way for solving the complex dilemma of choosing the appropriate averaging scheme of local scale behavior (measurement or parameters) for a correct transition from the local scale to the field scale behavior would be a comparison of measurement at different scales.

Unfortunately, this study was not a true comparison of measurements at different scales rather than a comparison of parameters estimation at two scales based on measurements at single locations. Specific design of experiments was prepared to determine the validity of estimating solute transport behavior at field scale as the average of all point measurements or the average of the local scale parameters over that spatial scale.

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