Local-scale solute transport in variously structured soils under continuous flood irrigation

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Abstract

This paper reports the results of research carried out to establish the possibility of using Time Domain Reflectometry (TDR) technology in measuring breakthrough curves of anion tracers such as chloride and bromide. The test conditions adopted refer to the case of undisturbed porous media with bimodal porosity under saturated-unsaturated conditions and to a liquid phase of assigned chemical characteristics. The BTCs were measured with vertically installed TDR probes. Interpretation of signal impedance was carried out in a similar fashion to Kachanosky et al. so as to obtain estimates of the minimum impedance load Z_0. The measured BTCs were interpreted to estimate transport parameters by using a two-parameter convective-dispersive model. Finally, a sensitivity analysis of the hydrodynamic dispersion parameter as related both to the average pore water velocity v, and TDR measurement errors were performed.
Keywords: solute transport models, chloride breakthrough curve, TDR calibration.

1 Introduction

Contamination of groundwater has received considerable attention in recent years because of concerns about degradation of this important source of fresh water, with possible toxicological consequences. Contamination has often been associated with point sources such as disposal sites or illegal dumping of chemical and industrial wastes. More recently, non-point pollution has become the center of increased attention by the research community, both in urban and

agricultural landscapes. Although a vast amount of complex mathematical flow and transport models exist, reliable predictions of flow and transport processes in natural, heterogeneous soils can only be made if expected spatial and temporal variability of the soil-atmosphere interface where diffuse pollution almost always occurs is also addressed. Unfortunately, the limiting step in our understanding of solute transport at field scale is just a lack of reliable and sufficient data. Indeed, at present investigations on solute transport in porous media for assessing solute leaching potential mainly base on time-consuming laboratory or field experiments (Kutilek and Nielsen, [2]; Jury et al., [3]).

Recent studies have demonstrated the potential of Time Domain Reflectometry (TDR) technology as an alternative technique to estimate chemical transport parameters in soil (Kachanosky et al., [1]; Comegna et al., [4]). This procedure could be recognized as an effective means for characterizing the solute transport behavior in soil.

Given the advantages of acquiring in real-time a suitable number of laboratory and field observations, which are particularly useful in parallel basic research, the Institute of Agricultural Hydraulics of the University of Naples "Federico II" together with DITEC of the University of Basilicata set up an equipment with a view to defining sufficiently and completely the parameters of chemical transport in soil for the widespread future use of simulation models. The main objective of this study is to illustrate the application of TDR methodology for estimating solute transport parameters from miscible experiments at laboratory scale. Main problems arising when applying the methodology to undisturbed soils with different aggregation degrees are also addressed. In the last part of the work results of modelling of experimental data are illustrated and a sensitivity analysis is also performed, mainly aiming to verify the robustness of fitted hydrodynamic dispersion parameter as related to experimental errors.

2   Experimental methodology

2.1 TDR in solute transport studies

A time domain reflectometry (TDR) waveform contains information describing both the speed of propagation and the attenuation of an electromagnetic pulse as it travels along a waveguide or probe (Topp et al., [5]; Spaans and Baker, [6]). Use of these properties allows for the determination of both the water content θ and the bulk electrical conductivity EC_a in approximately the same volume of porous medium (Dalton et al., [7]). Through long-established relationships describing the dependence of EC_a on θ and the electrical conductivity of the soil water EC_w, these two measurements promise the ability to determine EC_w from a TDR wave-form. For conditions under which the soil water chemistry is dominated by a single electrolytic solute, EC_w will be linearly related to the solute concentration, allowing for solute concentration measurement with TDR. Techniques for measuring the parameters of the transport models based on TDR have recently become widespread. Such techniques can be used to measure the mass flux of solute past the ends of TDR probes and characterize the probability

density function of solute travel time. The procedure may be applied to a conservative tracer under steady state leaching condition, provided that a solute pulse causing a measurable reduction in the TDR signal is added to soil surface.
As shown by various authors (Dalton and van Genuchten, [8]; Nadler et al., [9]; Kachanoski et al., [1]) the resident solute concentration C_r (ML^{-3}) in the soil may be determined from estimates of bulk soil electrical conductivity EC_a (dSm^{-1}), conducted by means of TDR.

In general, in a porous medium under conditions of steady flow, there are the following relations between the resident concentration C_r, bulk electrical conductivity EC_a and impedance Z_∞ (ohm), hereafter referred as Z, of the transmission line measured by TDR:

$$C_r = a + bEC_a \tag{1}$$

$$EC_a = \frac{K}{Z - Z_c} \tag{2}$$

in which a and b are calibration constants, K (L^{-1}) is the cell constant of the TDR probe and Z_c is the impedance associated to the cable, the connector and the tester (Topp et al., [5]).

Eqn. (1) holds for variable soil salinity levels ranging between 0 and 50 dSm^{-1} and for different values of θ (Ward et al., [10]), even if at higher θ values some nonlinearity was observed for low concentrations. In fact, EC_a is linearly related to EC_w for a range between 1 and 20 dSm^{-1} (Rhoades et al., [11]). As was also shown, EC_w is linearly related to pore water solute concentration C_r for a similar range. Thus the EC_a-C_r relationship is approximately linear for this electrical conductivity range (Kachanosky et al., [1]). The nonlinearity observed is held to be affected by the clay content in the soils and by Na$^+$ saturation, water content and probe size. Thus, the relation at low EC_w values, with θ constant, is soil-specific and should be estimated.

Direct calibration of the TDR probe in different solutions with known EC_w (0-12 dSm^{-1}) can be used to calculate Z_c and K (Heimovaara, [12]). For salinity levels less than about 3 dSm^{-1} $Z_c \ll Z_\infty$ (Mallants et al., [13]).

The relative concentration of the solute resident in the volume sampled by a TDR probe of length L is given by:

$$C_r^*(L,t) = \frac{C(t) - C_0}{C_0 - C_i} \tag{3}$$

equivalent to:

$$M^*(L,t) = \frac{M(t)}{M_0} \tag{4}$$

where C_0 is the concentration of the applied solution, C_i is the original concentration in the porous medium, $C(t)$ is the concentration for greater time values, M^* is the relative solute mass, $M(t)$ (ML^{-2}) is the total mass of solute between the pair of rods at any time t and M_0 is the total mass of solute contained between the rods from soil surface to the bottom end of the probe.

By substituting eqn. (1) in eqn. (3) and by using eqn. (2) to eliminate the constants, the following is obtained (Kachanoski et al., [1]):

$$M^*\left(L,t\right) = C_r^*\left(L,t\right) = \frac{Z^{-1}(t) - Z_i^{-1}}{Z_0^{-1} - Z_i^{-1}} \tag{5}$$

where Z_i is the initial impedance before applying the solution, Z_0 is the impedance associated with the applied concentration C_0, equation (5) shows that, with θ constant, the distribution $C^*(L, t)$ may be obtained from measurements of impedance $Z(L, t)$ if a particular value of impedance Z_0 can be related to a known concentration such as the concentration of the inlet solution C_0.

In a displacement experiment with a vertical TDR probe configuration, assuming that the TDR probe extends from the surface to a depth L, addition of a solute pulse with total specific mass M_0 at time t_0 immediately leads to a drop in impedance Z. When the finite solute pulse is wholly located in the volume observed by the TDR probe, the impedance will have a constant minimum value Z_0 associated to M_0. However, as the solute moves below the bottom end of the TDR probe, the impedance will tend to gradually increase and reach its initial value Z_i when all the solute is past the probe.

It is currently possible to obtain Z values directly from the display of a *Tektronix* (mod.1502C) cable tester. In this study, the readings were conducted at the intersection of the tester display cursor with the trace of the reflected signal when the width of the latter appeared stabilised. According to Nadler et al. [9], the problem of multiple reflections may be overcome by reading the impedance at a late time ($t\rightarrow\infty$) along the signal when all multiple reflections are suppressed. The differences in Z are used in the calibration process to obtain relative mass distribution (Kachanosky et al., [1]) from which in turn solute flux concentration at $z=L$ can be derived (Elrich et al., [14]).

By deriving eqn. (4) as regards time, the flow density relative to the solute may be obtained:

$$f(L,t) = \frac{1}{Z_0^{-1} - Z_i^{-1}} \frac{\partial}{\partial t}\left[Z(t)\right]^{-1} \tag{6}$$

Equation (6), equivalent to the probability density function (*pdf*) of solute travel time, may be considered as a characteristic of the volume of solute transport and may thus be assumed directly as a transfer function in the transfer function model *TFM* (Jury et al., [15]).

2.2 Miscible transport experiments

Methodology illustrated above was verified with reference to three undisturbed soil samples 150 mm in diameter and 170 mm in length from different sites of southern Italy. The samples were collected by means of steel samplers guided by an appropriate hydraulic device, ensuring the simultaneous removal of surrounding material so as to reduce alterations and compaction of the samples.

Samples were identified as *sandA*, *clay* and *sandB* respectively. Their main physical characteristics as determined in the laboratory, together with some

pedological characteristics of the horizons and the sampling site, are reported in Table 1 (Comegna et al. [4]).

In the laboratory the samples were saturated slowly from the bottom. Subsequently, the saturated hydraulic conductivity K_s was determined by the constant head method.

Table 1:   Physical characteristics of the soils examined.

Texture (ISSS)	Sand (%)	Silt (%)	Clay (%)	Bulk density (g/cm^3)	Organic matter (%)	Pedological classification
Sandy (sand A)	88.0	6.0	6.0	1.297	3.1	Horizon A: crumb structure; *entisol*
Clayey sand (clay)	39.72	25.43	34.85	1.240	1.9	Horizon A: polyedric-subang. structure; *vertisol*
Sandy (sand B)	80.0	12.0	8.0	1.285	2.1	Horizon A: granular structure; *andosol*

Subsequently, in a thermoregulated chamber (20±0.5°C), the samples to be characterised were subjected to very simple one-dimensional flow processes with a specifically constructed experimental leaching configuration. The leaching unit shown in figure 1 consists essentially of a soil column, a rain simulator, a vacuum unit and allows saturated and unsaturated flow experiments to be conducted.

The bottom end cap of the column supports a nylon cloth of 25 μmesh-wire with a bubbling pressure of ≈2.5 kPa. A bubble tower with a movable air entry tube allows the pressure potential h, of water to be imposed at the bottom of the sample. The column is then equipped with a vertically installed bifilary TDR probe for measuring the water content θ and impedance Z, and with two tensiometers for measuring the water potential h.

The laboratory-built TDR probe consists of two 5 mm diameter steel rods, 50 mm apart, 150 mm long extending from a perspex head enclosing a 1:1 matching *ferrite balun* (Spaans and Baker, [6]) and is connected to the measuring device by a 2m-long coaxial cable (RG 58U) with a characteristic impedance of 50 Ω. Porous fritted glass plate tensiometers (10 mm in diameter and with a bubbling pressure of ≈50 kPa) are inserted horizontally 5 cm and 15 cm down the soil column and are connected to pressure transducers. The leaching unit is completed by other basic components, namely: i) a Tektronix Mod. 1502C metallic TDR cable tester equipped with an RS 232 interface (Tektronix Inc.); ii) a personal computer for control, acquisition and data analysis; iii) a 50-needle (id. 0.6 mm) rainfall simulator; iv) a peristaltic pump serving the simulator; v) an automatic fraction collector to collect the effluent in small fractions (10-15 cc).
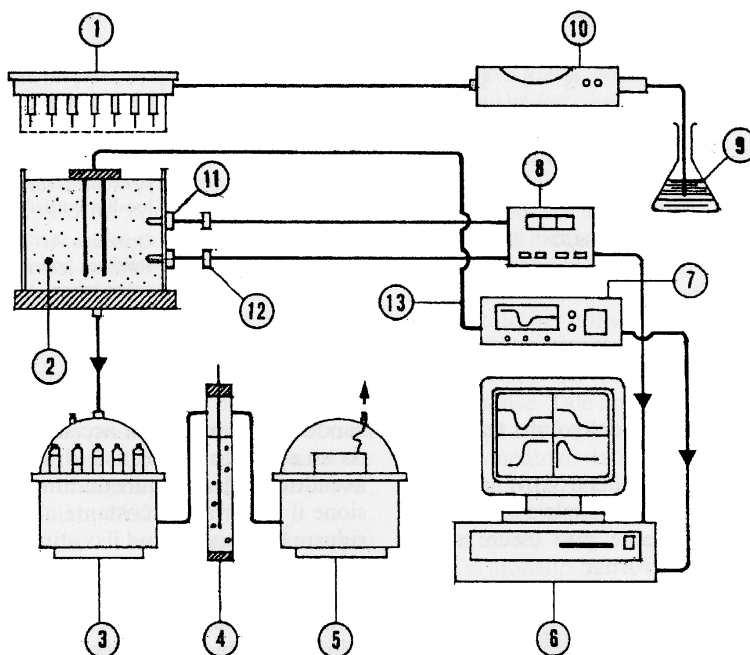
Figure 1:   Schematic diagram of the laboratory apparatus showing: (1) rain simulator, (2) soil monolith, (3) fraction sampler, (4) Mariotte vessel, (5) vacuum pump, (6) computer, (7) TDR tester, (8) multiplexer, (9) tracer vessel, (10) peristaltic pump, (11) tensiometer, (12) pressure transducer, (13) coaxial cable.

Each sample thus underwent a preliminary conditioning phase by feeding, with a steady water flux density, the flow system with a 0.01 N CaSO$_4$ solution until the steady flow and initial chloride concentration C_i=0 were reached. Contemporaneous measurements of θ, h and q allowed us to determine the onset of steady-state. At steady state, the solute was applied in a different way depending on the saturation degree during the experiments.

Concerning saturated experiments, the input of water was interrupted and a pulse of KCl of total specific mass M^* corresponding to 26.9 g/cm^2 of chloride, obtained by dissolving 1 g of KCl in 50 cc of water, was applied at the top of the sample for a variable period of time t_0 from 4 to 7 min. The chloride pulse, which had in the meantime completely penetrated the upper layers of the sample, was shifted by re-feeding the sample with the CaSO$_4$ solution.

During leaching tests the leachate was periodically collected. The chloride ion concentration at the outlet boundary was determined by titrating 10 mL of aliquot against standard AgNO$_3$ using 5% K$_2$CrO$_4$ as an indicator (=reference method) (Richards, [16]).

The $Z(t)$ time series were obtained and used to estimate by eqn. (5) the relative solute mass $M^*(L, t)$ localized between the surface and end of the TDR probe of length L.

3 Interpretative models of chemical transport

One-dimensional steady flow of an inert solute in homogeneous and isotropic porous media may be described by the classical convection-dispersion differential equation (CD):

$$R\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} - v\frac{\partial C}{\partial x} \tag{7}$$

where C (ML^{-3}) is the solution concentration, $v = q/\theta$ (LT^{-1}) is the average pore water velocity, q (LT^{-1}) is the Darcy velocity, θ the volumetric water content, D (L^2T^{-1}) the coefficient of hydrodynamic dispersion, R the retardation coefficient accounting for equilibrium linear sorption processes, x (L) is the distance in the direction of the flow and t (T) is time. Instantaneous concentrations C provided by TDR electrical resistance measurement are defined as the mass of solute per unit of soil volume C_r.

The hydrodynamic dispersion coefficient D has commonly been expressed as $D = \lambda v_0^n + D_0$, where λ and n are constants. λ is a characteristic property of the porous medium usually referred as dispersivity and n is commonly taken as a value between 1-2 (Kutilek and Nielsen, [2]).

On the basis of heuristic reasoning supplied by Jury et al. [3], equation (7) describes simply dispersion on a macroscopic scale in a homogeneous porous medium, hypothesizes implicitly that there is a Gaussian distribution of velocity in the flow domain and typically supplies a signal which may be modelled by a dispersion coefficient D in which all approximation errors are propagated.

During experiments some BTCs could show the establishment of a non-equilibrium indicated by tailing and early appearance of tracer in the effluent, mechanisms that generally cannot be described by analytical solutions supplied by equation (7). The mobile-immobile convection-dispersion concept (MIM) (van Genuchten and Wierenga, [17]; Gaudet et al., [18]) can be used for solute transport in such soils. In this conceptual model the porous medium is divided into two transport regions (*mobile* and *immobile*). Convective transport is assumed to occur only in the so-called *mobile* region, which is a fraction β, of the water filled pore space.

$$\theta_m \frac{\partial C_m}{\partial t} + \theta_{im}\frac{\partial C_{im}}{\partial t} = \theta_m D_m \frac{\partial^2 C_m}{\partial x^2} - \theta_m v_m \frac{\partial C_{im}}{\partial x} \tag{8}$$

$$\theta_{im}\frac{\partial C_{im}}{\partial t} = \alpha\left(C_m - C_{im}\right)$$

Chemical exchange between the two regions is assumed to be a diffusive process and is macroscopically described by a first-order kinetic, according to a difference of concentration, with a rate coefficient α.
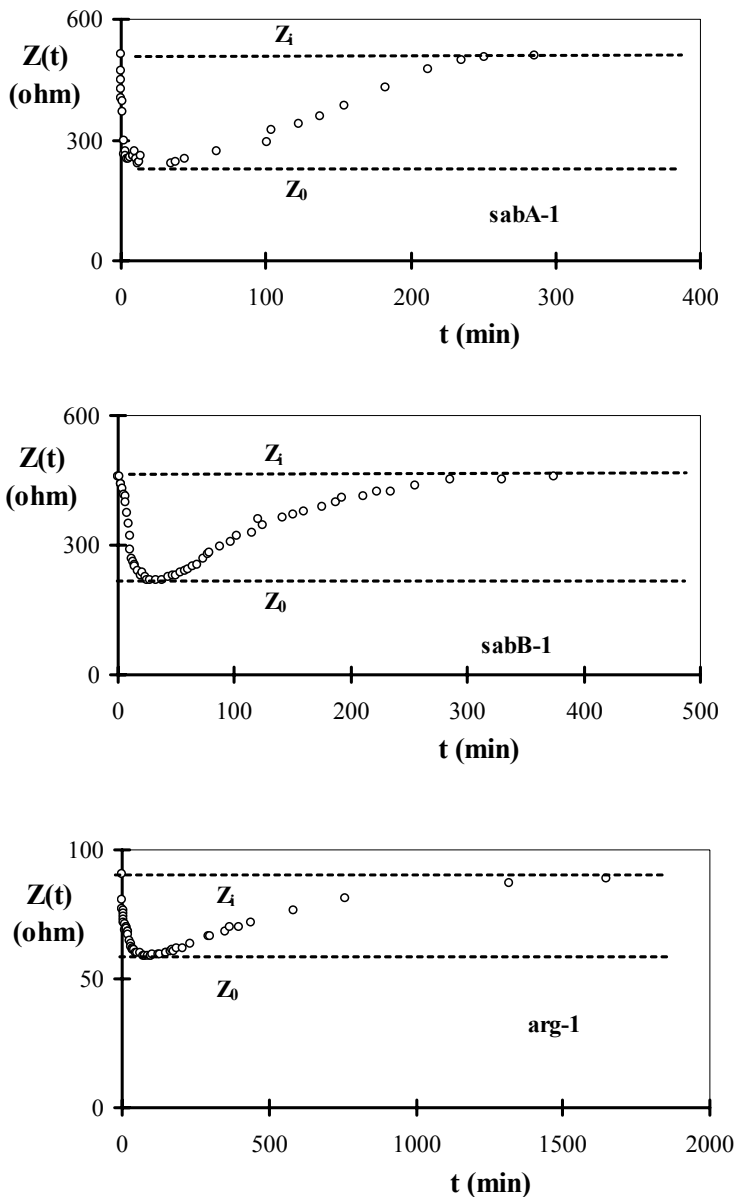

Figure 2: $Z(t)$ values measured by TDR.

4 Results and discussion

Overall, seven tests were carried out on *sandA* sandy soil, eight on *sandB* sandy soil and two on *clay* clayey soil. In order to illustrate the characteristic pattern of the electric impedence of transmission line in the soil, figure 2 reports as an example the time series of $Z(t)$ recorded during experiment *sandA*-1, *sandB*-1 and *clay*-1 conducted on respective columns at lclayest v_0 value and at complete saturation.

It may be observed from the figures that the initial impedance Z_i varies from 520 ohm for sandA-1 to 460 ohm for *sandB*-1 to 93 ohm for *clay*-1. Such differences may be attributed to the different chemical and physical characteristics of the soils examined.

Moreover, after application of the solute, significant variations are noted in impedance values, which in a short space of time record minimum Z_0 values of 250 ohm, 220 ohm and 58 ohm respectively for the three different soils. Finally, as shown by the times required to return to the Z_i values, the different physical and chemical characteristics of the soils also resulted in different time dynamics of solute transport.

The obtained values of Z_i, Z_0 and $Z(t)$ were re-used to plot the BTCs, $C(t)$, normalized between 0 and 1. For all the soils a straightforward comparison between TDR derived concentrations and those measured with the reference method may be established in figure 3 which represents the pertaining *pdfs*.

For *sandA-1* and *sandB-1*, the pdfs measured with TDR and the reference method are comparable to a slightly asymmetric bell-shaped curve. The results compare well, showing differences only for t>80min (r^2=0.960) and for t>60min (r^2=0.974). By contrast, for *vertisol*, the *pdf*s are typically asymmetric with accentuated tails (figure 4). A differentiation of the *pdfs* is noted, with clear deviations from the reference values, for times t<120min (r^2=0.882). In this case, the *pdf* measured with TDR leads to a slight overestimate of the time corresponding to the peak concentration and thus to a slight underestimate of the modal velocity of the solute.

Just as an example of results obtained under unsaturated conditions, figure 4 shows the diagram of $Z(t)$ values for the experiment *sandA*-2, along with the comparison between derived concentration values and those measured by the reference method.

The very slight discrepancies between the compared experimental BTCs emphasise the good agreement of the experimental devices and techniques adopted. For all the other experiments conducted, not reported here for the sake of brevity, it was observed that the initial impedence Z_i, corresponding to concentration C_i, varies within a range between 700 and 519 ohm. Such differences may generally be attributed to the variability in bulk soil electrical conductivity, the different water and clay contents of soils. After application of the solute, significant variations in impedance were noted which, with different time laws, showed minimum Z_0 values, corresponding to the applied concentration C_0, between 38-83 ohm.
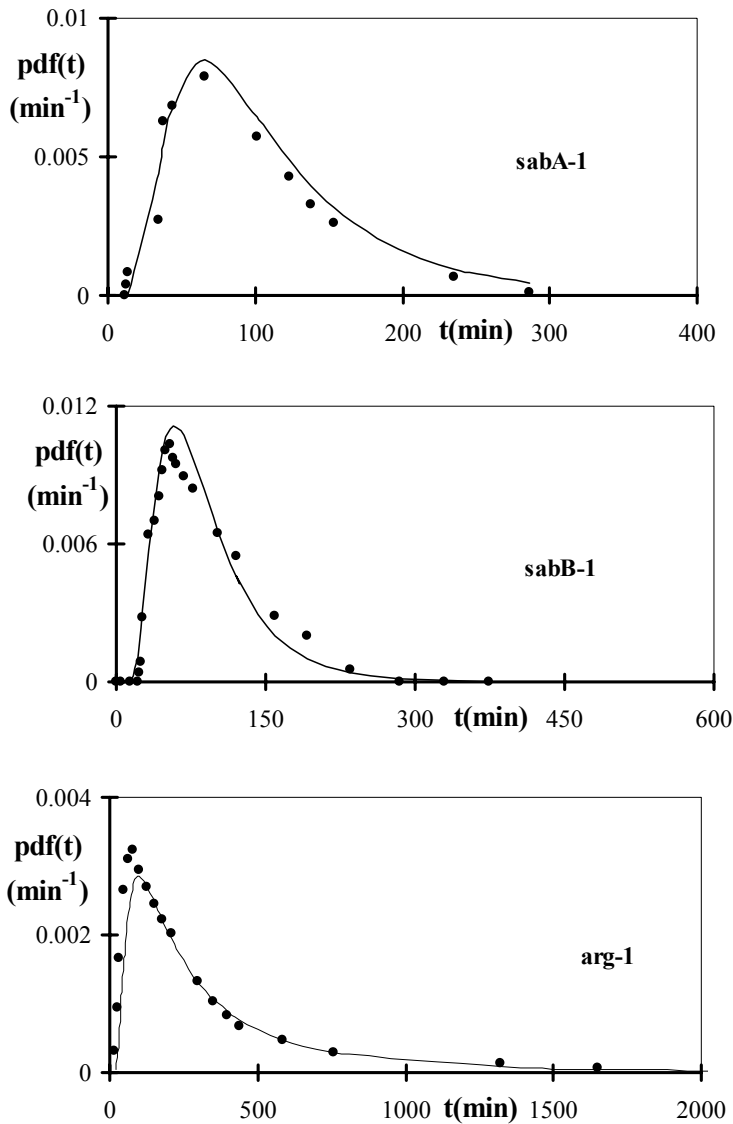
Figure 3:   Comparison between pdf(t) obtained by TDR (—) and pdf(t) obtained by the reference method (•).

   The experimental BTCs were first modeled with the CD model (equation (6)). The model parameters were determined using the CXTFIT program (Toride et al., [19]) which minimizes the sum of the squares of the residuals between the measured values and those calculated with an analytical solution of type A1

proposed by van Genuchten and Wierenga [17]. So as to reduce the number of iterations, the actual velocity v_0 was set equal to the q/θ ratio and initial estimates used of parameters R and D were reasonably close to optimal values, obtained with the deterministic methods proposed by Fried and Coubernous [20].
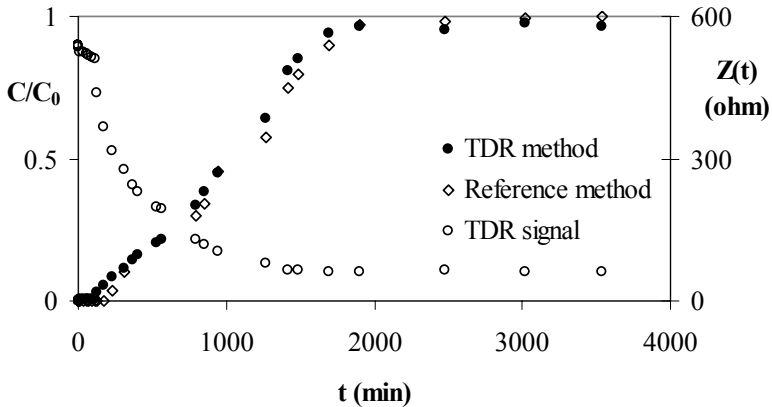

Figure 4:   Measured impedance load $Z(t)$ (*sandA-2 experiment*) from the cable tester (o); $C(x, t)$ values measured with TDR ($\bullet$) and reference method ($\Diamond$).

Table 2:   Parameters obtained by fitting the CD model to the experimental BTCs. R^* and D^* parameters starting values obtained by using the methods of van Genuchten and Wierenga (1976) and Fried and Combarnous (1971), respectively.

Parameters	Entisol	Vertisol	Andosol
q (cm min^{-1})	0.0715	0.0189	0.0732
Ks (cm min^{-1})	0.0640	0.0343	0.0343
θ_s	0.410	0.489	0.370
v (cm min^{-1})	0.174	0.039	0.198
R*	1.270	1.092	1.187
R	1.280	1.070	1.201
D*(cm^2 min^{-1})	0.418	0.184	0.658
D(cm^2 min^{-1})	0.275	0.146	0.451
P	9.512	3.962	6.580
λ (cm)	1.577	3.786	2.280
r^2	0.980	0.997	0.988
MSE	0.024	0.012	0.027

Seemingly a good agreement between calculated and measured BTCs was achieved, as confirmed by the high r^2 obtained also for experiments not shown here.

Though the shape of some breakthrough curves and pertaining parameters seemed indicate the establishment of a non-equilibrium, especially on columns labeled as *sandA* and *clay,* curve-fitting MIM model to experimental breakthrough produced always unimproved fittings and unreliable parameter estimates as compared to the CD results. Thus, deducting physical meanings from the uncertainly parameters obtained by MIM model would have been hazardous and probably meaningless. Accordingly, the obtained MIM parameters are not shown herein and all the following analyses were performed considering the CD as the appropriate model.

3.1 Sensitivity of hydrodynamic dispersion to experimental errors

Impedance Z, and average pore water velocity v, are the experimental parameters mainly involved in the hydrodynamic dispersion estimation. Any error in their measurement propagates on the dispersion coefficient in a way which can be studied through simulations.

Starting from an experimental curve for which the closest fitting was obtained, the sensitivity of hydrodynamic dispersion to errors in velocity determination was investigated by applying fictitious variations to its experimental value in the range -10%-10%. Velocity was first fixed while optimizing both dispersion and retardation coefficients. In this case a different hydrodynamic dispersion was obtained depending on the starting velocity value. To the contrary, a constant dispersion was obtained by optimizing both dispersion and velocity parameters, thus demonstrating the timeliness of optimizing velocity when unstable water content and/or inflow were hypothesized or observed during experiments. Results referring to these trials are observable in figure 5a and 5b.

Concerning the influence of impedance determinations on hydrodynamic dispersion, new fictitious experimental curves were calculated by perturbing the experimental $M^*(L, t)$ obtained from equation (5) through a random error which was calculated according to the following equation:

$$M_p^*(L,t) = M^*(L,t)\left[1 + 2f(F - 0.5)\right] \qquad (9)$$

where $M_p^*(L, t)$ is the relative solute mass which is assumed to be subject to random measurement error, F is a random number between 0 and 1.

Finally, f is the relative error to which values of 0.05, 0.10 and 0.15 are assigned in order to simulate errors of 5, 10 and 15% respectively. Parameter estimation analysis was carried out on the fictitious realizations of data using values shown in Table 2 for the experiment *sandA*-2 as starting values for transport parameters.

Graphical results are illustrated in figure 6. An appreciable sensitivity to measurement errors was observed just when the 15% error was considered, with a large deviation of D from its "true" value. Nevertheless, when 5 and 10% errors were applied, the observed deviations were limited to 1.1 and 2% respectively.
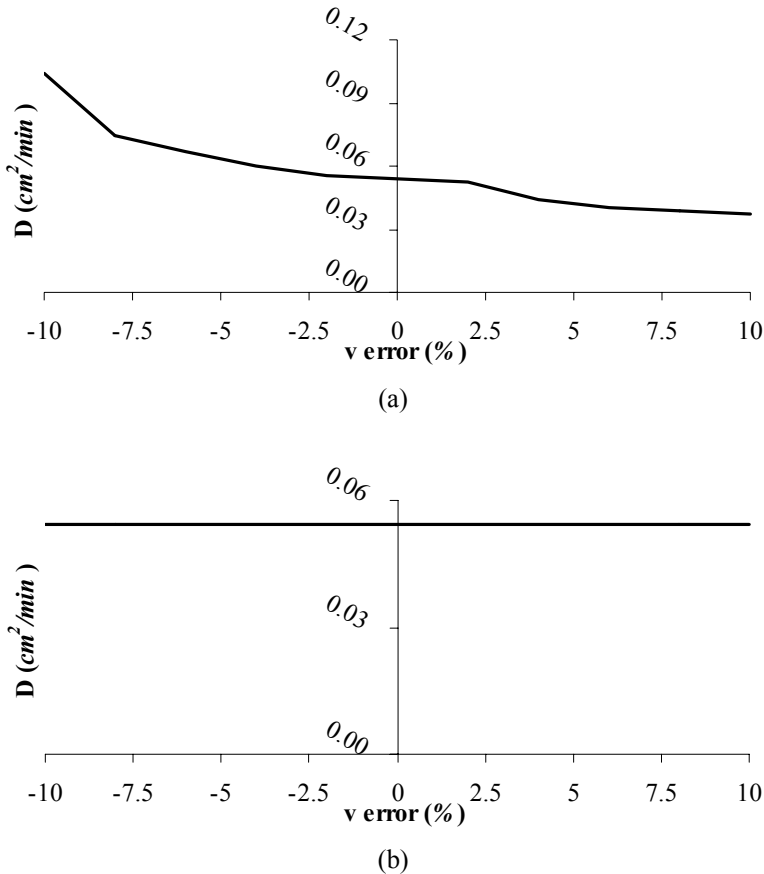
Figure 5: (a) Hydrodynamic dispersion coefficient as obtained by applying measurement errors to pore water velocity, keeping v fixed during the fitting; (b) Hydrodynamic dispersion coefficient as obtained by applying measurement errors to pore water velocity v is optimized during the fitting.

4   Conclusions

TDR technology, though already used in the field of soil physics and soil hydrology to measure water content in natural porous media, has only recently begun to be used to estimate chemical transport parameters in soil.

The illustrated methodology, which considers laboratory tests on undisturbed soil samples, permits considerable reductions in the number of measurements, while supplying results which are at least as accurate as those obtainable from long-established conventional techniques.
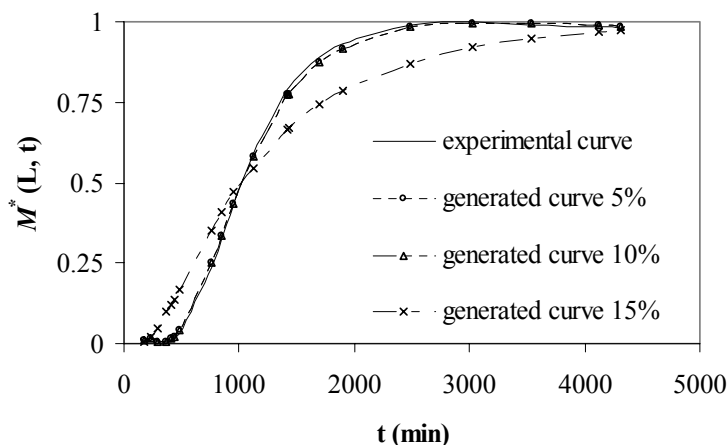
Figure 6:   Effect of errors in the determination of M^*(L, t) on the fitted curve.

Tested on soils with clearly different physical characteristics, the method allows reliable definition of transport parameters in steady flow conditions and for water content near saturation.

With regard to sandy soils, the two-parameter CD model supplied optimal estimates of concentrations, $C(x,t)$. As regards clayey soil, the use of the CD equation is not necessarily ruled out although a lower r^2 value was calculated. To reduce the greater discrepancies observed between the calculated curves and the measured data, more sophisticated versions of the CD model must be used, such as the CD-MIM model, which is known to compartmentalise the liquid phase into two domains, *mobile* and *immobile*. Nevertheless, CD appears a very reliable model which is appropriate for mechanistic of the solute transport process. Applying the technique to field trials may be extremely useful. In order to obtain sufficiently precise estimates of transport parameters for applications, techniques will have to be set up which take account of soil heterogeneity and the uncertainty of boundary conditions. Further and more extensive verification of the methodology is, however, necessary both in the laboratory and in the field, as well as on different soils.

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