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Using Surface Time Domain Reflectometry Measurements to Estimate Subsurface Chemical Movement

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Abstract

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Using Surface Time Domain Reflectometry Measurements to Estimate Subsurface Chemical Movement

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ABSTRACT

Chemicals that leach through soil pose threats to surface and groundwater quality. It is difficult and expensive to measure subsurface chemical transport and the transport properties required for extrapolating predictions beyond limited observations. The objective of our study was to evaluate whether solute transport properties measured at the soil surface could be used to predict subsurface chemical movement. The study was conducted in a greenhouse soil pit. The solute transport properties of the surface 2-cm soil layer were determined by using time domain reflectometry (TDR) to measure the bulk electrical conductivity during a step application of CaCl_2 solution. The movement of chemicals in the subsurface was measured within the top 30 cm of soil following a pulse input of CaCl_2 solution. A comparison of the measured chemical transport properties in the surface and subsurface zones of the soil showed that the parameters were similar. Furthermore, the estimated parameters determined by the surface TDR method were used to predict the chemical concentration distributions within the 30-cm soil layer, and it was found that the centers of mass of predicted chemical distributions were not significantly different from the measured ones. Therefore, the surface TDR measurements could be used to successfully predict subsurface chemical transport within the upper 30 cm of the soil. This surface measurement technique is a promising tool for vadose zone chemical transport studies.

SUCCESSFUL PREDICTION of the fate and transport of solutes in the subsurface hinges on the availability of accurate transport parameters. Most methods available for measuring chemical transport properties in the field are time-consuming and often result in extensive soil disturbance. Moreover, it is difficult and expensive to measure subsurface chemical transport and transport properties required for extrapolating beyond limited observations. Several numerical and analytical solute transport models are available for predicting chemical leaching, yet testing of the models in heterogeneous soil is limited due to a lack of observations.

Time domain reflectometry is a tool that can be used to obtain solute transport data (Kachanoski et al., 1992; Mallants et al., 1994; Vanclouster et al., 1995; Ward et al.,

1995). It is a method that enables nondestructive repeated sampling at specific depths in soil. Time domain reflectometry rods can be installed horizontally or vertically to obtain data to evaluate solute transport models in heterogeneous soils. Horizontal positioning of TDR probes enables the sampling of a relatively large soil volume perpendicular to vertical flow. This is useful for estimating solute fluxes in undisturbed field soils that exhibit small-scale heterogeneities due to the presence of macropores, immobile water regions, or zones of low-permeability (Mallants et al., 1996). However, it is not feasible to install horizontal probes at deep soil depths. Studies with horizontal probes were either limited to lysimeters or were preceded by extensive digging of soil trenches around the sites (Ward et al., 1995; Vanclouster et al., 1995; Mallants et al., 1996; Vanderborght et al., 2000). Digging to gain access for probe installations causes unwanted disturbance to the measurement sites. There exists a need to develop a technique for determining subsurface chemical transport with minimal labor and soil disturbance.

Vertically installed TDR probes provide a way to obtain data for various soil layers without causing major soil disturbance. However, a major limitation in using vertically installed TDR probes to study solute transport is the need for soil or layer specific calibration equations that relate signal attenuation to the resident concentration of an ionic tracer (Mallants et al., 1996; Vogeler et al., 1997). To simplify the calibration of TDR probes, Mallants et al. (1996) recommended using the most accurate and feasible measurements of TDR breakthrough curves (BTCs) obtained from the surface 5-cm layer of soil, where it was possible to reach equilibrium with a step input of tracer in a reasonable time. Deeper depths may require an inordinate amount of tracer solution and time to ensure proper equilibrium.

Lee et al. (2000, 2002) measured bulk electrical conductivity during a step input of tracer solution with TDR probes installed diagonally into the surface 2 cm of 20-cm-long undisturbed soil columns. They also measured effluent flux concentrations from the bottom of the soil columns. They analyzed their data with the two domain mobile-immobile (MIM) solute transport model developed by Coats and Smith (1964) and van Genuchten and Wierenga (1976, 1977). The MIM model divides soil water (θ) into two domains: a mobile water domain (θ_m) where water and chemicals move with mean pore velocity (V_m) and an immobile water domain (θ_{im}) where water is stagnant and chemicals move by diffusion only. Dispersion of chemicals takes place in the mobile domain and is similar to that in the convective-dispersive

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Abbreviations: CDE, convective-dispersive equation; MIM, mobile-immobile [model]; TDR, time domain reflectometry.

equation (CDE). The water in the immobile domain is connected to water in the mobile domain and allows for chemical diffusion between the two domains. The MIM model is written as follows:

$$\theta_m \frac{\partial C_m}{\partial t} + \theta_{im} \frac{\partial C_{im}}{\partial t} = \theta_m D_m \frac{\partial^2 C_m}{\partial z^2} - q \frac{\partial C_m}{\partial z} \quad [1]$$

where, C_m and C_{im} are the concentrations of chemicals in the mobile and immobile domains ($M L^{-3}$), D_m is the dispersion coefficient ($L^2 T^{-1}$) in the mobile domain, q is the flux density ($L T^{-1}$), t is time (T), and z is depth (L). Chemical transfer between the two domains is proportional to the concentration difference between the two domains and can be described as a first-order process (van Genuchten and Wierenga, 1976):

$$\theta_{im} \frac{\partial C_{im}}{\partial t} = \alpha (C_m - C_{im}) \quad [2]$$

where α is a first-order mass exchange coefficient (L^{-1}).

The MIM model has been successful in describing numerous column experiments and explaining tailing on BTCs (Nielsen et al., 1986). Lee et al. (2002) determined one set of MIM solute transport properties from surface TDR measurements and a second set of MIM properties from the effluent data. They found similar surface and column transport properties. Furthermore, they were able to use the MIM transport parameters derived from the TDR surface measurements to accurately predict the column effluent concentrations. One-dimensional flow was used in the Lee et al. (2002) column studies, and it is not clear whether similar TDR measurements can be used to accurately determine surface solute transport properties under two- or three-dimensional flow conditions. Further testing of the Lee et al. (2002) surface TDR method for determining solute transport properties is warranted.

The main objective of this study is to extend the Lee et al. (2002) surface TDR method to a three-dimensional flow condition. The TDR method is used to determine surface solute transport properties. The properties are evaluated by how well they predict subsurface leaching. Experiments are performed in a greenhouse soil pit containing disturbed soil. Although the soil is disturbed, it is not homogenized. The soil in the pit has been cropped for several years, and the soil surface has been tilled. The disturbed soil experiment is used as a step toward evaluating and refining this method for future application in undisturbed field soil.

METHODS AND MATERIALS

Site Description

The experiment was conducted in a greenhouse soil pit. The soil in the pit had clay loam texture (0.34 sand, 0.42 silt, and 0.24 clay mass fraction). The soil pit was about 60 cm deep and was underlain by sawdust and undisturbed soil (Jaynes et al., 1995). The soil pit was tilled before conducting the experiment. The surface transport properties were measured by a dripper-TDR experiment followed by the measurement of subsurface leaching with a ponded infiltrometer experiment.

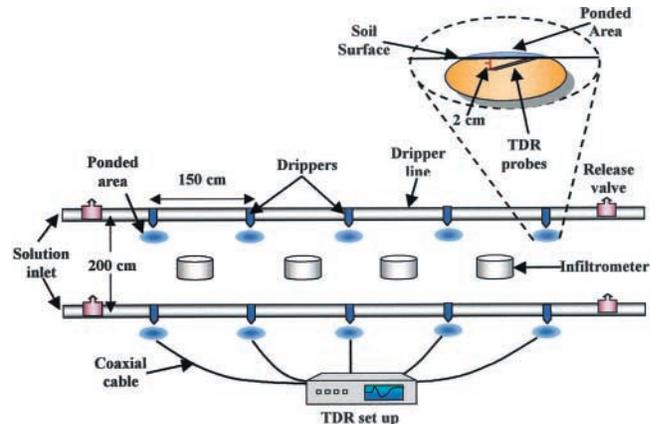


Fig. 1. Schematic diagram of the experiment.

Dripper-TDR Experiment for Surface Measurements

Al-Jabri (2001) and Al-Jabri et al. (2002) introduced a dripper irrigation system as a point source of water or solute to determine the hydraulic and solute transport properties of shallow soil in situ at multiple locations at the same time with minimal disturbance. We used a similar dripper irrigation system along with TDR probes to determine surface chemical properties in the soil pit (see Fig. 1). The setup included multiple drippers as point sources of solutes and TDR probes to measure the bulk electrical conductivity of soil. The dripper irrigation tubing was equipped with five drippers spaced at 1.5 m. Each dripper had a designed discharge rate of $4 L h^{-1}$ within the applied pressure range (pressure-compensating drippers). The dripper irrigation tubing was placed at two different positions on the surface of the soil pit, providing for a total of 10 surface measurement locations.

The TDR set up consisted of two-rod, 3.8-mm-diam, 100-mm-long probes, a cable tester (model 1502B, Tektronix Corp., Redmond, OR), and a computer program to store and analyze the data. The TDR probes were inserted at an angle from the surface to a depth of 2 cm to minimize soil disturbance. For the steady-state leaching experiment with a step increase of input solute concentration, relative solute concentration $R(t)$ can be represented as (Lee et al., 2000):

$$R(t) = \frac{C(t) - C_i}{C_0 - C_i} = \frac{ECa(t) - ECa_i}{ECa_0 - ECa_i} \quad [3]$$

where C_i is background solute concentration, C_0 is input solute concentration, ECa_i is TDR-measured EC for C_i , and ECa_0 is TDR bulk EC corresponding to C_0 . Under steady-state conditions, we can directly use ECa values to determine solute transport properties in soil. Because of the linear relationship between ECa and C , the empirical constants do not need to be determined to calculate $R(t)$. In this study the real time electrical conductivity, $ECa(t)$ was determined with the aid of the Win TDR99 (Or et al., 1998) computer program.

It was assumed that each TDR probe measured the average bulk soil electrical conductivity of the soil surrounding the probe. A background steady-state condition was attained by applying 0.005 M $CaCl_2$ solution through the drippers until the EC readings and ponding radius reached constant values. Once the steady-state condition was attained, 0.2 M $CaCl_2$ solution was applied by the same drippers as a step input tracer for long enough time to allow the input solution to move deeper than 2 cm. $ECa(t)$ of the soil surface was measured continuously by the TDR setup.

After applying the tracer solution for about 2 h, soil samples from the surface 2-cm layer were collected from each TDR

probe location to determine the actual resident chemical concentration of soil solution, C . Each soil sample was split into two subsamples. One subsample was used to determine the gravimetric water content, and the other subsample was diluted five times with water. The diluted subsample was shaken and allowed to settle. The supernatant was then centrifuged at 9200 g for 20 min. The water from the sample was subsequently analyzed for EC with a conductivity meter (Model 30, Accumet, Hudson, MA). Knowing the final soil water solution concentration, $C(t)$, input tracer concentration, C_0 and background concentration, C_i , it was possible to determine the final value of the relative resident concentration, $R(t)$ in Eq. [3]. Subsequently, the corresponding final value of ECa_0 in Eq. [3] was determined and was applied to normalize the $R(t)$ with respect to the real time $ECa(t)$ values. The normalized $R(t)$ values represented the relative resident concentrations of the surface 2-cm soil layer, where the TDR probe was installed. The relative resident concentration BTCs obtained from TDR were used to estimate the MIM solute transport parameters, θ_m , α , and D_m . Preliminary analysis showed that θ_m determined by the Clothier et al. (1992) method and inverse curve fitting of one-dimensional pore velocity within TDR probe regions, V_m , α , and D_m provided the best fit of observed TDR data. Mallants et al. (1994) also pointed out that the fitted (optimized) pore velocity V_m was more appropriate than the measured one because the TDR only samples a small region of the whole flow domain, and the region might or might not include preferential flow channels.

Subsequently, the mobile fraction θ_m/θ was determined by Clothier et al. (1992) method, which was equal to the final relative resident concentration of the soil samples collected after infiltrating $CaCl_2$ solution. In the CXTFIT program (Toride et al., 1999), the value of θ_m/θ was fixed and V_m , α , and D_m were determined by inverse curve fitting of the TDR BTCs for each location. The depth for the curve fitting in CXTFIT was set at 1 cm, which was the average depth of the sampling volume (0–2 cm) of the TDR probe.

Ponded Infiltrometer Experiment to Measure Subsurface Leaching

Ponded infiltrometers were used to apply a pulse of $CaCl_2$ solution for use in the subsurface chemical transport study. Following the dripper–TDR surface measurements, infiltrometers were installed at the center of two adjacent dripper locations. A total of four cylinder infiltrometers with inner diameter of 45 cm and height of 25 cm were inserted 10 cm deep into the ground (see Fig. 1). With each infiltrometer, a steady infiltration rate was established by ponding tap water at a depth of 2 cm for 4 to 5 h. Once steady infiltration was achieved, the tap water supply was stopped, and after the ponded tap water infiltrated, 5 cm of 0.2 M $CaCl_2$ was applied uniformly via a sprinkler can inside each infiltrometer. As soon as the 0.2 M $CaCl_2$ solution infiltrated in each infiltrometer an additional 2 cm of tap water was applied. Immediately after the infiltration of the tap water, soil samples were collected at four locations in each infiltrometer. To avoid soil compaction, the top 5 cm of soil was sampled with 5.4-cm-diameter rings before sampling the 5- to 30-cm layer as a 2.54-cm-diameter core. The collected samples were immediately divided into approximately 3-cm sections to determine the distribution of solute concentration (EC) within the soil profile. Soil solution extracted from each section was analyzed for EC.

The relative resident concentrations for the soil profile were used in the inverse curve fitting program of CXTFIT to determine the subsurface MIM parameters. Since the Clothier et al. (1992) method for determining θ_m/θ was not applicable in this

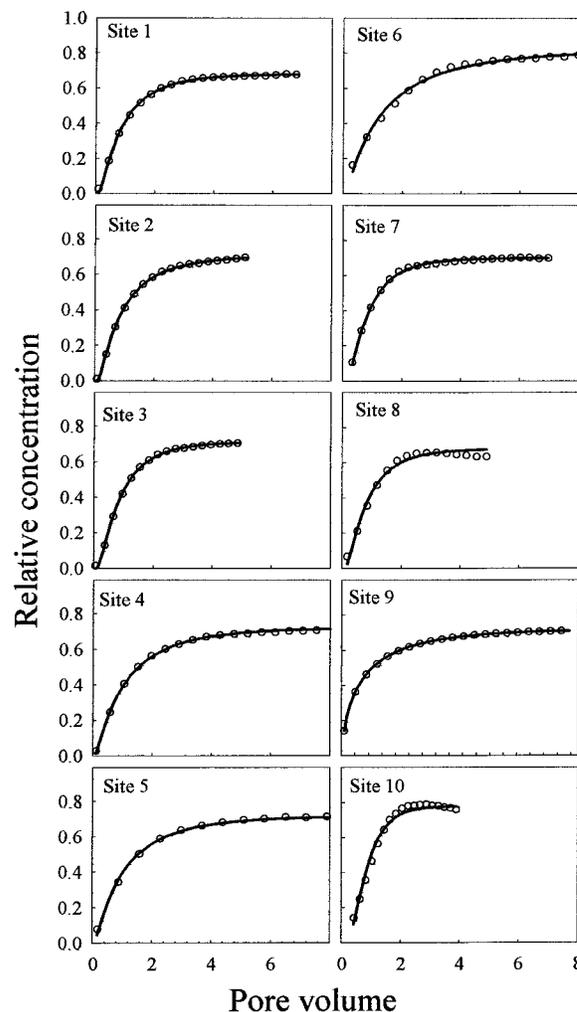


Fig. 2. Observed relative resident concentration for the surface soil determined by dripper–TDR experiment. The circles and solid lines are the measured and fitted values, respectively.

pulse input experiment, all of the subsurface MIM parameters were estimated by inverse fitting the observed resident concentration distributions. However, to minimize the number of fitted variables, the measured subsurface pore velocity, V_m , was used. Therefore, CXTFIT was used to estimate θ_m/θ , D_m , and α . In addition, the surface solute transport properties determined by the dripper–TDR setup were applied using the same pulse input as in the ponded infiltrometers in the direct mode of the CXTFIT program to predict soil profile resident concentration profiles. To compare the estimated surface and subsurface transport properties, a statistical test was performed. The statistical test was a nonparametric, Wilcoxon–Mann two-samples test that is suggested for data sets with unknown distributions obtained from different processes (SAS Institute, 1996). Moreover, the center of mass obtained from the predicted profile BTCs were compared with the observed center of mass within the soil profile.

RESULTS AND DISCUSSION

Surface vs. Subsurface Transport Properties

The observed resident concentration BTCs obtained by TDR measurements for the soil surface layer are shown in Fig. 2. Soon after the application of chemical tracer,

Table 1. Comparison of surface and subsurface solute transport properties.

Parameters	Surface method	Subsurface method	Statistical comparison
Number of observations	10	16	
Pore velocity, V_m , cm h^{-1}	10 (± 3) [†]	30 (± 2)	***
Dispersion coeff., D_m , $\text{cm}^2 \text{h}^{-1}$	10 (± 5)	40 (± 22)	***
Immobile water fraction, θ_{im}/θ	0.28 (± 0.02)	0.31 (± 0.06)	NS [‡]
Mass exchange coeff., α , h^{-1}	0.04 (± 0.06)	0.20 (± 0.15)	NS
Dispersivity, λ , cm	1.02 (± 0.4)	1.28 (± 0.68)	NS
Center of mass, cm	13.5 (± 0.81)	13.8 (± 0.62)	NS

*** Significant at 0.001 probability level.

[†] Values in parentheses are confidence intervals at 0.05 probability level.

[‡] NS, nonsignificant at the 0.05 probability level.

the increase in resident chemical concentrations was relatively fast, but with time, the rate of increase reduced and approached an asymptotic value. Conceptually, in the beginning the mobile water (or active flow pathways) was replaced by the input tracer solution mainly due to convection, resulting in a sharp increase in the resident concentration, whereas during the latter process the increase in resident concentration was slow because the chemical exchange process between mobile and immobile domain occurred by diffusion. At most of the locations, the resident concentrations approached constant values, thus indicating negligible diffusion.

The applied surface emitter discharge rates for all 10 drippers ranged from 3.9 to 4.3 L h⁻¹. The relative resident concentrations from the soil extracts ranged from 0.68 to 0.82, indicating the presence of an immobile water domain. Subsequently, the average θ_{im}/θ determined by Clothier et al. (1992) method was found to be 0.28 (± 0.02). The θ_{im}/θ was then fixed during the inverse curve fitting of the TDR data to determine V_m , D_m , and α . The mean V_m , D_m , and α determined by CXTFIT were, respectively, 10 ± 3 cm h⁻¹, 10 ± 5 cm² h⁻¹, and 0.04 ± 0.06 h⁻¹ (Table 1). The coefficients of determination, R^2 , for the inverse curve fitting by CXTFIT program were 0.98 or greater for all of the sites, indicating good fitting of the observed data.

The subsurface properties were determined for a total of 16 locations by sampling four 30-cm-deep soil cores in each of the ponded infiltrometers installed at four different locations between the dripper locations. The relative resident chemical concentration distributions for the soil profiles are shown in Fig. 3. As a result of a pulse input of CaCl₂, the chemical distribution within the soil profile varied with depth, with the peak relative resident concentration occurring at an average depth of about 13 cm. The average maximum relative resident concentration was 0.69 ± 0.06 . The percentage of chemical mass recovery within the 30-cm soil profile ranged from 69 to 115% with an average value of 90%, indicating some movement of chemical below the 30-cm depth at most sites. However, we were successful in capturing a substantial amount of tracer within the sampled portion of the soil profiles. All three MIM parameters θ_{im}/θ , α , and D_m were obtained by fitting the observed relative resident concentration distributions. The average θ_{im}/θ , α , and D_m for the soil profiles were found to be 0.31 ± 0.06 , 0.20 ± 0.15 h⁻¹, and 40 ± 22 cm² h⁻¹, respectively (Table 1). The average θ_{im}/θ for surface soil and soil profiles were similar; however, the confidence intervals for

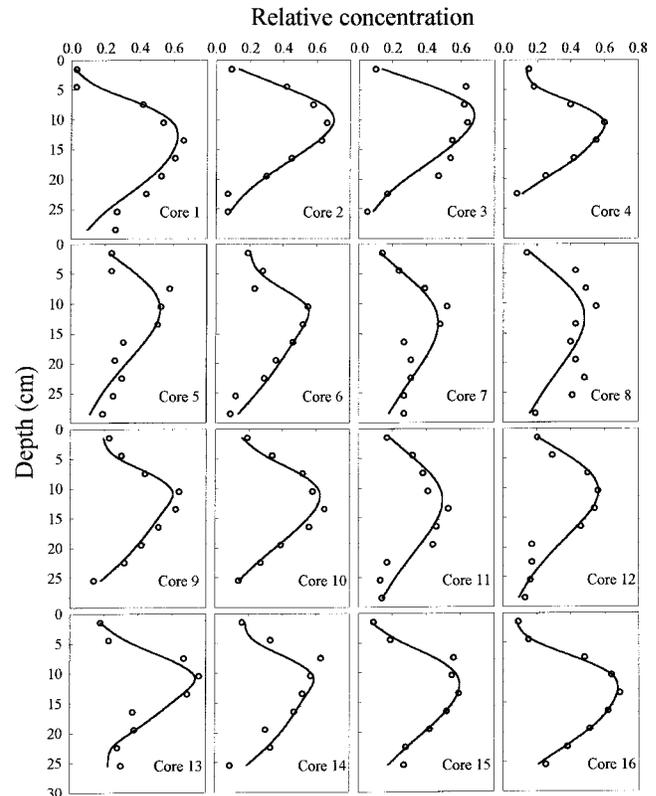


Fig. 3. Observed relative resident concentration for the subsurface soil measured from the ponded infiltrometer experiment. The circles and solid lines are the measured and fitted values, respectively.

soil profile parameters were comparatively large. Individual α values and their corresponding confidence intervals were greater for the soil profiles than for the surface soil layers. The coefficient of variation of the α values was larger for the surface soil than for the soil profile. In a majority of the cases, the value of α was zero for the surface soil. The dispersion coefficient, D_m , was lower at the surface than in the soil profile, which was due to the corresponding lower pore velocity. The average measured subsurface pore velocity in the ponded infiltrometer experiment was 30 cm h⁻¹, which was significantly larger than the fitted estimated one-dimensional pore velocity at the surface dripper-TDR locations (10 cm h⁻¹). Therefore, to eliminate the effect of different pore velocities, we used dispersivity ($\lambda = D_m/V_m$) as the quantitative measure of dispersion, and it was found that both at the surface and in the soil profile, the dispersivities were similar with average values of 1.02 ± 0.4 and 1.28 ± 0.68 cm, respectively (Table 1). Using the nonparametric test (Wilcoxon-Mann two-samples test), no significant difference was found between the MIM parameters, θ_{im}/θ , α , and λ measured at the surface and in the soil profile. These findings agree with the results from Lee et al. (2002), where the values of θ_{im}/θ , α , and D_m obtained by the surface TDR method were similar to the parameters estimated from undisturbed soil column effluent data. Furthermore, both surface and subsurface measurement methods met the flow rate and cumulative infiltration requirements as suggested by Snow (1999) to obtain accurate estimates of transport parameters.

Predicted vs. Observed Resident Concentrations

The surface transport properties determined by the dripper–TDR setup were used to predict the subsurface resident concentrations for 30-cm-deep soil profiles. We used center of mass as the quantitative measure for comparing predicted and observed resident concentrations, and it was found that the difference between the two centers of mass was not significant. The average centers of mass from predicted and observed BTC were found to be 13.5 ± 0.81 and 13.8 ± 0.62 cm with coefficients of variation of 9 and 7%, respectively (Table 1). We also conducted a nonparametric test (Wilcoxon–Mann two-sample test) that can be used for data sets with unequal numbers of observations and found that the difference between centers of mass was not significant. These are promising results indicating the capability of the surface TDR method to provide solute transport parameters that can be used to extrapolate chemical movement into deeper soil.

SUMMARY AND CONCLUSIONS

A study was performed in a greenhouse soil pit to test whether surface measurements of chemical transport properties could be used to accurately predict subsurface chemical transport. TDR was used to determine the transport properties for the surface soil, while soil profile resident chemical concentration distributions were analyzed to determine subsurface transport properties. The surface and subsurface chemical transport properties including immobile water fraction (θ_m/θ), mass exchange coefficient (α), and dispersivity (λ) were found to be similar. Furthermore, the surface transport parameters obtained from the dripper–TDR setup were successful in predicting the center of mass for the subsurface resident concentration distributions. The TDR method is relatively simple and requires only a surface soil sample with minimum disturbance of soil, after applying a step input of salt solution. So far, the technique has been tested in undisturbed columns and in a disturbed soil pit. Further testing and refinement of the method under a range of soil conditions is needed. This surface measurement technique is a promising tool for vadose zone chemical transport predictions, and the technique may prove to be useful for assessing subsurface chemical leaching under different management practices.

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