

## **Apparent Transport Parameters Determined Using Three Conservative Tracer Applications**

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## Abstract

The dispersivity ( $\alpha$ ) component of mechanical dispersion is typically assumed to be a characteristic of a soil and independent of soil water pore velocities. Mechanical dispersion ( $D_m$ ) is assumed to dominate molecular diffusion ( $D_0$ ) when estimating hydrodynamic dispersion ( $D_{hd}$ ). Two conservative tracers were applied at three separate times: bromide in 1992 and 2000 and chloride in 1995. Tracer breakthrough was monitored using twenty-six passive capillary samplers (PCAPS). Breakthrough curves were fit to solutions to the Advection Dispersion Equation (ADE) to calculate transport parameters. On average,  $\alpha$  values were 3.74, 20.6, and 16.1 cm for the 1992, 1995, and 2000 applications, respectively. There were no significant correlations between  $\alpha$  values and calculated pore water velocity for individual samplers. Dispersivities at a given location did not show a correlation in time. Spatially,  $\alpha$  values significantly fit a log-normal distribution. PCAPS calculated could not predict observed  $\text{Br}^-$  profiles generated through soil sampling because PCAPS  $\alpha$  values did not reflect tracer spreading caused by heavy rainfalls during the second half of the breakthrough. These findings show a weakness in the current conceptual model describing  $D_m$  as dependent on a static  $\alpha$ . Differences between cycles of molecular diffusion and mechanical dispersion dominated periods were more important to determined solute dispersion than were values of  $\alpha$ .

## 1. Introduction

Estimating realistic parameter values with predictive capacity is a challenge when modeling natural systems. For solute transport models within porous media, engineering estimates of parameters are often based on easy to measure characteristics such as the hydraulic conductivity and particle size of the media. The Advective Dispersion Equation (ADE) is a common model used to simulate solute transport in porous media because it has few parameters.

To apply the ADE, estimates of hydrodynamic dispersion ( $D_{hd}$ ) must be made. Hydrodynamic dispersion is the sum of molecular diffusion ( $D_0$ ) and mechanical dispersion within the system. Mechanical dispersion ( $D_m$ ) is the product of the dispersivity ( $\alpha$ ) and the pore water velocity ( $v$ ). Dispersivity is considered to be a characteristic parameter of the soil media [2,17], and therefore should be independent of the pore water velocity [21].

Researchers have attempted to describe the relationship between  $\alpha$ , other parameters in the ADE, and the soil environment. Studies within deep groundwater and saturated soils have shown a positive correlation between  $\alpha$  and the scale length of tracer travel [8,12]. Variations in  $\alpha$  seem to be less understood in unsaturated conditions where measured  $D_{hd}$  and  $\alpha$  have not been well-correlated with depth [16]. Wierenga and Van Genuchten [21] found no increase in the dispersion with depth in unsaturated water columns. A slight decrease of  $\alpha$  with depth was found under drip and ponded irrigation [13]. Persson and Berndtsson [15] studied the effect of water application on transport parameters and found that the  $\alpha$  decreased when water was applied more often. This relation was attributed to less variability in the pore water velocity with frequent water applications.

For unsaturated conditions, Beven et al. [2] compiled an extensive review of dispersion coefficients,  $\alpha$  values and pore water velocities obtained from tracer studies of various area and length scales. In general,  $\alpha$  values have fallen between centimeters for packed column experiments to many meters in deep groundwater studies.

Numerous studies have recorded the importance of preferential flow within soil systems [11,12,14,18,20]. Soil systems showing preferential flow exhibit much different chemical transport properties than soils showing predominately flow through the soil matrix. Studies have found that preferential flow can deliver nitrate ( $\text{NO}_3^-$ ) and/or pesticides to shallow groundwater at short transport times. Hamdi et al. [11] recorded separate peaks for the preferential and matrix components of the tracer breakthrough. Water can follow preferential flow paths during times when water inputs exceed the infiltration capacity and saturated conditions develop. Due to the spatial heterogeneity of hydraulic conductivity (K) and infiltration (I), saturated conditions may be localized and difficult to spatially characterize. Kung et al. [14] observed early tracer breakthrough due to preferential flow though no saturated conditions or ponding was observed.

A majority of tracer studies showing preferential flow flooded the soil surface or used irrigation systems to apply water at rates nearing the infiltration capacity of the soil and thus selected for conditions that favored preferential flow. These observations may be applicable to other irrigated sites, sites with low permeability soils, and areas with short duration, high intensity rainfall events such as in the Midwest. It is likely that these results do not represent the processes in many other regions with different rainfall patterns or well-drained soils.

Because of the high velocity and short travel times observed in studies using saturated and or continuous irrigation sets, it is plausible that flow characteristics and fitted dispersion parameters could differ from experiments relying on natural rainfall. For example, time of peak tracer breakthrough is typically shorter than the time that would be needed under natural conditions.

Due to the high pore water velocities employed in many experiments,  $D_m$  has been much greater than  $D_0$  within the system, to the point where  $D_0$  could be assumed to be negligible. Mathematically,  $D_{hd} = D_0 + \alpha V$  is reduced to  $D_{hd} = \alpha V$ , where the product of  $\alpha V$  is  $D_m$  [19]. However, if one is concerned with the behavior of the slowest moving solutes, such an assumption is not acceptable [10]. In soil systems where pore water velocities are variable, with times of slow movement and some preferential flow, diffusion into soil peds may be important [20].

It is likely that diffusion would be important in natural systems experiencing cyclic rainfall throughout the year such as Oregon's Willamette Valley. During periods of little or no water movement solutes could move by diffusion through the smaller water-filled soil pores. The effect of such conditions would be a long tail on the tracer breakthrough and may be expected to have high fitted  $D_{hd}$  depending on the measured concentrations within the breakthrough tail relative to the concentration within the bulk tracer breakthrough [10].

Several studies have pointed out the log-normal distribution of spatially measured transport parameters within soil systems. Boll et al. [3] used wick and gravity samplers to measure conservative tracer breakthrough under a variety of soil types and water application rates. Dispersion, pore water and solute velocities best fit a lognormal distribution under macropore dominated systems, but fit a normal distribution under matrix flow conditions. Under a week of continuous flooding and sampling using suction cup samplers, Jaynes et al. [12] found that the pore water velocity and dispersion of a  $Br^-$  tracer best fit a lognormal distribution. Saturated conditions and preferential flow through macropores was also observed. Gupte et al., [9] measured  $Br^-$  breakthrough from drainage tiles and found a lognormal distribution for measured dispersivities.

The objectives of this study were to use wick samplers and soil sampling to examine: 1) the characteristics of tracer breakthrough for three pulse applied conservative tracers under natural rainfall conditions, 2) the relation between fitted transport parameters for each of the applications under predominately matrix flow conditions where  $D_0$  is important 3) the spatial distribution of fitted  $\alpha$  values 4) the ability of the ADE to spatially predict tracer concentration.

## **2. Methods**

### **2.1 Site Description**

The study site was at the North Willamette Research and Extension Center (NWREC) located near Aurora, Oregon ( $45^\circ 17' N$  and  $122^\circ 45' W$ ). The Willamette Valley has a Mediterranean climate characterized by wet winters and dry summers. Weather records

from 1963 to 1990 give an annual average precipitation total of 103.6 cm (40.8 inches) (Oregon Climate Service, 2003). During the dry, sunny summer months, potential evapotranspiration (ET) far exceeds rainfall.

Soils at the 0.98 hectare study site are slightly sloped towards the south (<3%). The soils are of glaciolacustrine genesis and are classified as a Woodburn variant loam with a small strip of Willamette variant loam bisecting the field. Description of organic carbon, pH, bulk density, particle size distribution, saturated hydraulic conductivity and moisture retention characteristics for the NWREC study plot are described in Brandi-Dohrn et al. [4].

Field management with respect to soil tillage remained the same over the study period. There was evidence of burrowing rodents at the site, most commonly in the border areas between the experimental plots. Efforts were made to control the rodents with varying success. The rodents may have changed conductivity and bulk density values both spatially and temporally. The southeast corner of the field had a slight depression and often became ponded in the winter. Soil sampling here showed a compacted layer at 0.7 m depth with low hydraulic conductivity.

Summer vegetables grown on the experimental plots were irrigated using a hand move sprinkling system. Irrigation sets took place once per week. Water was applied for variable set times according to the time of the year; however a quantitative model of evapotranspiration was not used to fine tune irrigation amounts. Irrigation procedures were consistently managed by the same person throughout the entire study.

## **2.2 Field Lab Methods**

Passive Capillary Samplers (PCAPS) were used to continuously sample soil water flux. Twenty-six PCAPS were located throughout the 0.98 ha field. A detailed description of PCAPS design, installation and operation at the NWREC is detailed in Brandi-Dohrn et al. [4].

Water was pumped from the PCAPS after cumulative rainfall between sampling events neared 2.5 cm, at which point the collection vessels would be reaching capacity.

The procedures for PCAPS generated sample processing and flow weighting calculations are provided in Feaga, [6].

Water content was measured during the summer of 2002 using a neutron probe model 503-DR Hydroprobe from Campbell Pacific Nuclear International. The unit was calibrated from nine access tube sites throughout the field, and measurements were made at 17 sites in total. Access tubes were located approximately 2.4 m from the center of the PCAPS. For the calibration, soil cores were taken at 10 cm increments down to 160 cm depth and neutron probe signal was compared with laboratory measured water contents. Soil water content was determined in the field 6 times and measurements were always made the day before irrigation events.

Conservative tracers were applied over the lysimeter plots three times during the 11 year study. The first  $\text{Br}^-$  tracer was applied on the 4<sup>th</sup> of November, 1992. A  $\text{Cl}^-$  tracer was applied on the 28<sup>th</sup> of October 1995. The final  $\text{Br}^-$  application took place on the 12<sup>th</sup> of December, 2000. In order to remain consistent during the study, 29.6 g/L  $\text{Br}^-$  and  $\text{Cl}^-$  solutions were used for each tracer application. The solutions were prepared by dissolving KBr or KCl in water. Each of the tracers was applied to the field using a three meter wide sprayer mounted to a bicycle wheel. The solution was pressurized using  $\text{CO}_2$  and applied at 414 kPa. A 0.5 mm depth of the solution was sprayed above each pair of PCAPS from a height of one meter. The first two applications covered a 3 m x 7.5 m area. Due to poor tracer mass recovery obtained from the first two applications, the last  $\text{Br}^-$  tracer spray area was doubled to 6 m x 7.5 m. The assumption being that the larger tracer area would minimize the effect of lateral solute movement on tracer recovery.

Following the Dec 12<sup>th</sup>, 2000 application of the  $\text{Br}^-$  tracer at the NWREC, manual soil sampling was used to observe spatial concentrations of the  $\text{Br}^-$  plume. Dates for the three sampling events were the 5<sup>th</sup> and 6<sup>th</sup> of May 2001, 5<sup>th</sup>-7<sup>th</sup> of September 2001, and the 15<sup>th</sup> of December 2001, respectively. For each sampling event, two holes were augured (1 ¼" diameter) on each of the subplots equipped with working PCAPS. All augured holes were well within the area of tracer application and about 1.5 meters from the buried samplers.

The last two sampling events were identical to the first except that each 1.2 meter hole was divided into twelve layers (instead of six), each 10 cm deep. This change was made so that the Br<sup>-</sup> plume and resident concentrations of NO<sub>3</sub><sup>-</sup>-N within the soil water could be determined with better resolution. The upper 30 cm were not represented during the September sampling event due to large quantities of decaying bean crop left from the recent mowing and a dry, uneven soil surface that was deemed difficult to characterize using an auger sampling tool. Samples were not taken from fallow plots containing PCAPS 21 and 22 in both September and December. In September the soil was too compacted to sample, while in December the area was ponded with water.

Soil Br<sup>-</sup> and NO<sub>3</sub><sup>-</sup> were extracted from each of the soil samples using the method used by Dick and Tabatabai [5].

### 2.3 Advective Dispersion Equation

The advection dispersion equation (ADE) with linear adsorption / exclusion was used to model observed breakthrough curves of the three tracer applications. The differential form of the ADE for a tracer moving through the soil is:

$$\frac{1}{R} \frac{\partial C}{\partial t} = D_{hd} \frac{\partial^2 C}{\partial z^2} - v_w \frac{\partial C}{\partial z} \quad (1)$$

Here, concentration as a function of time and space is controlled by both advection and dispersion. A one-dimensional vertical solution to the ADE for a surface application whose mass is as a delta-function at  $x = 0$  (infinitely concentrated at the surface) is:

$$C(z,t) = \frac{M}{\theta AR \sqrt{4\pi D_{hd} t / R}} \left[ \exp\left(-\frac{(z - \bar{v}_w t / R)^2}{4D_{hd} t / R}\right) + \exp\left(-\frac{(z + \bar{v}_w t / R)^2}{4D_{hd} t / R}\right) \right] \quad (2)$$

Where:

- $C(z,t)$  = Concentration of tracer in soil water as a function of time and depth  
 $M$  = Mass of tracer applied over sampling area  
 $\theta$  = Volumetric fraction of soil occupied by water, taken to be field capacity 0.344  
 $A$  = PCAPS horizontal flux area  
 $D_{hd}$  = Hydrodynamic Dispersion Coefficient (encompasses both molecular diffusion and dispersion)  
 $R$  = Dimensionless retardation coefficient  
 $z$  = Stationary point of observation (sampler depth)  
 $\bar{v}_w$  = Mean pore water velocity

The semi-infinite form of the model utilizing two superimposed advecting Gaussian solutions effectively accounts for the fact that there is no flux with time due to diffusion out of the soil surface into the atmosphere.

The ADE was fitted to measured tracer concentrations using Microsoft Excel's Solver package to minimize the sum of squared differences between measured data and the modeled solution. Three ADE parameters were fit to the measured data:  $M$ , mass of  $\text{Br}^-$  added at time zero,  $D_{hd}$ ,  $R$ , the retardation coefficient. It was necessary to fit  $M$  to use the ADE but the fitted value was not imperative to describe the spreading of the solute or the temporal aspects of the center of mass. It was necessary to fit  $R$  because assigning a default value of one would force all breakthrough peaks to occur at one pore volume. A breakthrough at one pore volume would theoretically be expected with a conservative tracer in homogeneously packed porous media. Observations of tracer breakthrough did not support this. Fitting an  $R$  value can help describe solute behavior in systems showing preferential flow, unstable wetting fronts, and ion exclusion.

Unlike modeling situations where conventional time units can be used, such as the case of an advecting solute in a river with constant velocity, we used time units of relative pore volumes passed. This was necessary because solute movement for each of the tracers was driven by natural precipitation. Due to unsteady dry and wet conditions, the solute remained in the soil column for long periods of time with no movement. To

calculate the depth of water equivalent to exactly one pore volume, P.V., at a depth of the PCAPS,  $z$ , we used.

$$P.V. = z\theta_f \quad (3)$$

Where:

$\theta_f$  = Field capacity or volumetric fraction of soil occupied by water, measured at 0.344.

The P.V. was a constant value of 41.3 cm throughout the experiment. A unitless parameter applicable specifically to the field site, Relative pore volumes, p.v., and was calculated using:

$$p.v. = \frac{\sum_{i=1}^n V / A}{P.V.} \quad (4)$$

Where:

$V$  = Cumulative volume of water collected during sampling events

$n$  = Number of sampling events since tracer application

Hydrodynamic dispersion ( $D_{hd}$ ) is the cumulative effect of all processes causing the spread of solutes in porous media and can be expanded as follows to show the contribution of molecular diffusion ( $D_0$ ) and mechanical dispersion ( $D_m$ ).

$$D_{hd} = D_0 + D_m \quad (5)$$

Where:

$$D_m = \alpha \bar{v}_w \quad (6)$$

Where:

$\alpha$  = Dispersivity

The pore water velocity was not fitted, but was calculated using the cumulative volume of water collected during the period that breakthrough characteristics were measured for each of the tracer pulses. Pore water velocity could be calculated for any desired time increment,  $t$  (from average velocity over entire breakthrough to maximum velocity during periods of high rainfall and percolation):

$$\bar{V}_w = \frac{\left[ \sum_{i=1}^n V/A \right] \frac{1}{\theta_f}}{t} \quad \text{Or} \quad \bar{V}_w = \frac{p.v.*z}{t} \quad (7) \text{ and } (8)$$

Where:

$n$  = Number of sampling events in period of interest

The sum of least squared differences method was not capable of isolating  $D_0$  and  $D_m$  during the fitting process. The fitting process was only able to fit an effective spreading of the tracer plume and generate a value for  $D_{hd}$  by using the pore water velocity and fitting an intermediate dispersivity ( $\alpha^*$ ) for the system.

$$D_{hd} = \alpha^* \bar{V}_w \quad (9)$$

Where:

$\alpha^*$  = Intermediate fitting parameter reflecting the dispersivity in the system

The intermediate parameter was mathematically indiscriminate of the contribution of spreading due to  $D_0$  and  $D_m$ .

Conventional time was eliminated from the ADE using pore volumes and the total distance of the soil column. The use of pore volumes instead of conventional time provided a linear scale for the time variable required to fit the ADE to observed data.

$$D_{hd}t = \alpha^* \bar{V}_w t \quad (10)$$

$\bar{V}_w t$  = distance wetting front has traveled =  $z p.v.$

The real dispersivity of the system (as well as the mechanical dispersion) was calculated using the diffusion coefficient for  $\text{Br}^-$  adjusted to account for the inherent tortuosity in the soil media. An estimate of the unitless tortuosity coefficient,  $k$ , is 0.4 [1]. This is nothing but an algebraic rearrangement of Equation 5 and 6.

$$\alpha = \frac{D_{hd} - (D_0^* k)}{\bar{V}_w} \quad (11)$$

$D_0^*$  = Diffusion coefficient for  $\text{Br}^-$  with tortuosity constant  
 $k$  = Tortuosity coefficient

#### 2.4 Spatial Analysis Using the ADE

Chemical concentration is a function of time and space. Manual soil sampling provided an opportunity to test the ability of the ADE to predict the spatial chemical concentration profiles at a point in time. Transport parameters were calculated using the spatial concentration profile (spatial parameters) and compared with PCAPS calculated parameters (temporal parameters). Both the May 2001 and September 2001 sampling dates provided a concentration profile encompassing the rising and falling limb of the breakthrough curve. Therefore these dates could be used to generate unique solutions using the advection dispersion equation. The December sampling event took place during the time of or after the  $\text{Br}^-$  peak concentration had passed the 1.2 meter depth, therefore, the profile could not be used for analysis because it could not provide a unique set of solution parameters with only the falling limb of the tracer breakthrough.

To fit the parameters of  $\alpha$ , initial solute mass, and retardation, the cumulative number of pore volumes passing through the 1.2 meter flux plain up to the soil sampling date had to be determined using the PCAPS. These values, used as the time variable in the ADE,

were not very different for the day 143 and day 280 (May and September) of 2001 soil sampling events as little water was collected by PCAPS between these months.

### 3. Results

#### 3.1 Tracer Breakthrough and Transport Parameters Measured Using Stationary PCAPS

Tracer breakthrough was measured using each of the 26 PCAPS for each of the three tracer applications. Bromide concentration was analyzed following the 1992 Br<sup>-</sup> application for 47 sampling events between 4/12/1992 and 1/11/1994. Chloride concentration was analyzed following the 1995 application for 43 sampling events between 4/11/1995 and 20/4/1997. The final Br<sup>-</sup> application of 2000 was analyzed over 73 events between 19/12/2000 and 26/4/2003. Rainfall patterns during the time following the application of each of the tracers varied with respect to monthly and total accumulation (Figure 1). Months recording high precipitation had more periods of intense rain than low rainfall months.

The neutron probe provided water content with depth throughout the 2002 growing season to early fall (Figure 2). Measurements were made before weekly irrigation sets. The soil column was driest at the surface and wettest at 100 to 140 cm. Water content observations did not rule out the possibility of short periods of upward water movement caused by tension forces. Upward water gradients would make solute travel distances greater, allowing for more solute spread and longer travel times. Possible upward water gradients do not interfere with the study goal to characterize  $\alpha$  in field soils, as all plots were comparable under similar water contents.

The ADE was fit to all 78 breakthrough curves generated during the study. For each of the tracer applications, a flow-weighted average concentration for all 26 samplers was calculated and plotted against the average number of pore volumes collected. This provided an average breakthrough curve for each of the tracers (Figure 3 a,b, and c).

Differences were observed between the timing and shape of the three average breakthrough curves. The average breakthrough generated from the 1992 Br<sup>-</sup> application had the latest time of peak breakthrough and its shape had little skew. For breakthrough

curves measured through times at a point in space, the magnitude of skew is proportional to the amount of hydrodynamic dispersion in the system. The 1995  $\text{Cl}^-$  application resulted in the fastest time to peak breakthrough and had the most positively skewed fitted concentration curve, indicating that the plume was the most dispersed. The 2000  $\text{Br}^-$  application average breakthrough curve was skewed slightly less than the 1995 tracer application.

Average  $\alpha$  values for the three tracer applications, calculated from the flow-weighted average breakthrough curves, were 3.7 cm, 20.6 cm and 16.1 cm for the 1992, 1995, and 2000 events, respectively. Dispersivity values for all tracer applications ranged between 0.02 cm and 79 cm. The distribution of fitted dispersivities for each tracer application is shown in a boxplot (Figure 4).

Peak tracer concentrations reached sampler depth after 0.91, 0.6, 0.79 pore volumes or 404, 95, and 367 days for the 1992, 1995, and 2000 events, respectively. Average retardation ( $R$ ) values, defined as the ratio of the water velocity over the solute velocity, were 0.98, 0.74, and 0.83 for the 1992, 1995, and 2000 events, respectively. Pore water velocities were  $1.2 \times 10^{-2}$ ,  $2.2 \times 10^{-2}$ , and  $2.0 \times 10^{-2}$   $\text{cm h}^{-1}$ , for the 1992, 1995, and 2000 applications, respectively. Average transport parameters and their ranges for each of the tracer applications are summarized in Table 1, and include the parameters calculated through soil sampling discussed further in the text.

Average mass recovery in percent for the 1992  $\text{Br}^-$ , 1995  $\text{Cl}^-$ , and 2000  $\text{Br}^-$  applications were 32, 45, and 41% respectively. Differences in  $\text{Br}^-$  collection efficiencies for the 2000 tracer application below fallow and cover cropped plots were 47 and 35 %, respectively. Winter cover crops had a significant effect on collection efficiency at  $P = 0.021$ . The difference may have been due to scavenging and retention by the winter cover crop [6]. Cover crop treatment did not have a significant effect on  $\alpha$  values (Table 2). Though cover crops removed 12 % of the  $\text{Br}^-$  mass relative to the fallow plots, the attenuated  $\text{Br}^-$  pulse under cover cropped plots was still able to generate transport parameters comparable to fallow plots.

### 3.2 Relationship of Rainfall, Pore Water Velocity, and Molecular Diffusion on Tracer Breakthrough

Tracer application year was the only variable having a significant effect ( $P$ -value < 0.0001) on  $\alpha$  over the course of the three tracer applications. ANOVA results are given in Table 2. The PCAPS used for these measurements were stationary in space and management on the vegetable plots was constant throughout the study. It is unlikely that soil structure below the plow layer changed significantly in the relatively short 11 year study period. By deduction, rainfall patterns are the only remaining variable that could have affected the difference in measured transport parameters between years.

Many studies observe early tracer peak concentrations that are contributed to preferential flow paths in the soil. This soil hydrologic system appeared to be dominated by matrix flow, although evidence of preferential flow existed at the site. Steady rising and falling concentrations were observed for most breakthroughs. Observations of early-arriving highly concentrated tracer spikes were not widespread for the 78 breakthroughs.

Despite the predominance of matrix flow, preferential flow was an important flow regime at the site. Observations from individual samplers during the 1995 Cl<sup>-</sup> application breakthrough showed early arriving tracer. During the rainy December of winter 2001, several intense storms caused ponding on the soil surface. In seven days, from the 11<sup>th</sup> to the 18<sup>th</sup> of December, almost 8 cm of rain fell. Water was sampled from PCAPS four times during this period. Analysis of Br<sup>-</sup> concentration revealed peaks and troughs in the breakthrough curve between 0.6 and 1.2 pore volumes passed. Before the storm events, Br<sup>-</sup> concentrations were beginning to reach their fitted peak concentration of about 16 mg L<sup>-1</sup> (Figure 3c). Saturated conditions subsequently lowered Br<sup>-</sup> concentrations to about eight mg L<sup>-1</sup> as the ion was washed from locations near the preferential flow paths and dilution ratios were raised [20]. After the intense rain events, Br<sup>-</sup> concentrations returned to about 14 mg L<sup>-1</sup> as matrix flow once again dominated.

Providing other evidence of preferential flow, during near freezing mid-winter rain events in 2001, some samplers supplied “new” water that was considerably colder to the touch than water pumped from other samplers during the same event. Typically, temperatures in December and January are around 4°C (40°F) while soil temperatures are

higher, so water using complete soil matrix flow paths would be expected to have temperatures moderated by higher soil temperatures before reaching sampler depth.

Pore water velocity values were compared with calculated  $\alpha$  values to reveal possible correlations. A linear correlation (r-value 0.99) existed between the average pore water velocities calculated for the 1992, 1995 and 2000 tracer applications and their corresponding  $\alpha$  values. There was no correlation, however, between pore water velocities and  $\alpha$  values when individual sampler-generated values were compared. Since the variable  $\alpha$  values calculated for each tracer application could not be explained by average pore water velocity, these results suggest that  $\alpha$  values are more likely a function of cumulative monthly rainfall, rainfall timing and rainfall intensity. Average  $\alpha$  had only an indirect correlation to pore water velocity due to the associative relationship of these parameters.

Two breakthrough curves measured by individual PCAPS during the 1992 and 1995 application breakthroughs are shown in Figure 5 a and b. These breakthroughs illustrate the general effect rainfall patterns had on tracer transport. Figure 5a shows the measured breakthrough for sampler number 17 from the 1992 Br<sup>-</sup> application. The breakthrough had showed very little hydrodynamic dispersion (represented by skew) and generated an  $\alpha$  value of 0.02 cm when  $D_0$  was considered (Equation 11). Though this breakthrough was an extreme example, all of the breakthroughs from the 1992 Br<sup>-</sup> application showed little  $D_m$  and a greater relative contribution of diffusion. Referring to rainfall characteristics in Figure 1, there was unusually low rainfall during the winter of application and during the second winter. Though short-term intensity values were not available for this period, average pore water velocities were considerably lower and less variable than values calculated for the other tracer periods. Much of the rainfall during the second year of the 1992 application served only to replenish the soil water storage reservoir in the column and thus did not contribute greatly to Br<sup>-</sup> movement.

The highly dispersed breakthrough measured by sampler number 6 during the 1995 Cl<sup>-</sup> application is shown in Figure 5b. It is clear that heavy rainfall, especially soon after tracer application, had a large effect on breakthrough shape and fitted transport parameters. This highly skewed breakthrough had a fitted  $\alpha$  value of 49.9 cm and was a

result of the heavy rainfalls (159% and 170% of the thirty year average during water years 1996 and 1997, respectively) received immediately after tracer application and during the following year. Highly concentrated  $\text{Br}^-$  was collected at 0.2 to 0.4 pore volumes passed, showing evidence of preferential flow. Average pore water velocities were the highest for this tracer application. Periods of preferential flow between periods of diffusion resulted in large dispersion values. Although peak concentrations occurred at 0.6 pore volumes,  $\text{Cl}^-$  continued to be collected at detectable concentrations even after 2.0 pore volumes had been collected.

Dispersivity values from the 2000  $\text{Br}^-$  application fell between the 1992 and 1995 tracer applications. Very little rain fell during the first winter following the  $\text{Br}^-$  application; however, the second winter received above average rain and included some intense rain events that caused some preferential flow. Evidence from soil sampling indicated that little  $\text{Br}^-$  dispersion occurred during the first winter, yet the second winter succeeded in spreading the  $\text{Br}^-$  and resulted in an average fitted  $\alpha$  value of 16.1 cm.

### 3.3 Peclet Number Analysis

The Peclet number provided a quantitative method to assess the relative contributions of  $D_m$  and  $D_0$  to overall observed tracer spread ( $D_{hd}$ ). The theoretical Peclet number is calculated as the ratio between the magnitude of the average pore water velocity,  $v_w$ , times a characteristic length,  $d$ , expressed at the mean grain size, to the  $D_0$  coefficient [1].

$$Pe = \frac{\bar{v}_w d}{D_0} \quad (12)$$

The characteristic length may be expanded to include other scalars, for example, the ped size of structured soils [1].

Bear [1] showed the relationship between  $D_{hd}$  and  $D_0$  as a function of the Peclet number and defined the graphical relationship by dividing the curve into several zones. As the zone number increased from one through five, the importance of  $D_0$  diminished.

In the vadose zone, conditions usually fall into zone two to three [17] and  $D_0$  and  $D_m$  are both important. Peclet number in zone two to three are between 0.4 and ten.

For this Peclet analysis, as in the calculation of the true  $\alpha$  (Equation 11), the rate of  $D_0$  for  $\text{Br}^-$  was adjusted for tortuosity by multiplying the published value for  $D_0$  in pure water by 0.4 [1]. This resulted in a  $D_0$  value of  $2.88 \times 10^{-2} \text{ cm}^2 \text{ hr}^{-1}$ . Using this rate of  $D_0$ , a grain size for silt of 0.001 cm, and an average pore water velocity calculated for the 2000  $\text{Br}^-$  application of  $0.02 \text{ cm hr}^{-1}$ , a Peclet number of  $6.9 \times 10^{-4}$  was calculated. This small Peclet number indicated that all spreading of the tracer plume should be attributed to  $D_0$ . Indeed,  $D_0$  was an important process for solute spreading, especially during times of low pore water velocity such as in the summer when the tracer was below the depth of irrigation inputs.

It is unreasonable to believe that  $D_0$  is the dominant process at all times throughout the year. During the winter, pore water velocities were the greatest and there was evidence of preferential flow during large rain events. The Peclet number increased to  $5.6 \times 10^{-3}$  during short periods (three to four days) when pore water velocities were higher due to large rain events. This Peclet number still remained too small for  $D_m$  to be an important spreading process. It appears that these results do not seem to well-describe solute spreading in this soil system, at least when we consider the fundamental silt grain size.

Below the plow layer, ped structures increase the characteristic diameter controlling solute spreading. If one assumes that the fundamental structure of the soil consists of peds with dimensions on the order of centimeters, Peclet numbers reach the magnitude where  $D_m$  is important. Using the average pore water velocity and a ped size of two cm or five cm gives Peclet numbers of 1.4 and 3.5, respectively.

Hydrodynamic dispersion ( $D_{hd}$ ) was fit during the least squares fitting process used to find  $\alpha$ . An experimental Peclet number could be calculated as shown in Equation 13.

$$Pe = \frac{D_{hd}}{D_0} \quad (13)$$

Experimental Peclet numbers from each of the tracer applications are shown in Figure 6. From this analysis we see that Experimental Peclet numbers were much higher than the theoretical Peclet number calculated with the average pore water velocity and the silt grain size diameter. It appears that tracer spread in this soil system was indeed determined by characteristic length scales on the order of cm's and not by the fundamental silt grain size. With the greater range of Peclet numbers calculated experimentally, solute spread would result from both  $D_0$  and  $D_m$  processes.

### 3.4 Spatial and Temporal Distribution of Fitted Dispersivity Values

Spatial and temporal variability of  $\alpha$  within porous media was analyzed using the 78 fitted breakthrough curves. In order to make each set of 26 dispersivities fitted from each tracer application comparable, the  $\alpha$  values were normalized by dividing each sampler's fitted  $\alpha$  by the mean  $\alpha$  calculated from all 26 samplers during each tracer event:

$$\alpha_{normalized} = \frac{\alpha_i}{\bar{\alpha}_j} \quad (14)$$

Where:

- $i$  = Dispersivity generated by one sampler during one tracer breakthrough
- $j$  = Tracer application (1992, 1995, 2000)

Normalizing removed the effect of environmental conditions such as tillage, rainfall, and pore water velocity on  $\alpha$  values. Normalizing transformed all dispersivities to a distribution with a mean of one.

Dispersivities measured by individual samplers were compared to reveal whether values remained constant over time in relation to the other samplers. It is traditionally supposed that the  $\alpha$  of porous media is a property of the soil and therefore should not change in time in absence of physical disturbance [2,17]. First,  $\alpha$  data sets for each of the tracer applications were paired by sampler number and the correlation coefficient between annual values was calculated. The dispersivities of the three tracers showed no

correlation with correlation coefficients of -0.003, 0.020, and 0.175 for comparison of Br 1992 vs Cl 1995, Br 1992 vs Br 2000, Cl 1995 vs Br 2000, respectively.

It appeared that areas of the field measuring extremely low or high  $\alpha$  values may have exhibited more consistent  $\alpha$  values. Sampler 17 had the lowest  $\alpha$  for all of the tracer applications. The number 6 sampler had the highest  $\alpha$  during the 1995 Cl<sup>-</sup> application and the second highest  $\alpha$  during the 2000 Br<sup>-</sup> application, but measured an average  $\alpha$  during the 1992 Br<sup>-</sup> application. These observations were likely attributed to soil structure. The consistently low dispersivities measured by sampler 17 for all applications could be due to a well packed homogenous soil with little secondary structure to promote preferential flow. Flow through soils above sampler 17 would be expected to be entirely through the soil matrix because they showed very little chemical spread in time. Figure 4.5a shows the 1992 Br<sup>-</sup> tracer breakthrough from sampler 17, which corresponded well to these expectations. In contrast, higher dispersivities similar to those fitted by sampler number 6 from the 1995 Cl<sup>-</sup> application (Figure 5b) could be due to some established preferential flow paths within the soil.

Statistical hypothesis t-tests were another method to identify any temporal correlation between fixed position samplers. Fitted dispersivities from the three tracer pulses were compared by determining whether a particular location consistently fit dispersivities above or below the mean. The null hypothesis was that the sample of three normalized  $\alpha$  values calculated by each PCAPS came from a population with a mean value of one. The alternative hypothesis was that the  $\alpha$  values came from a population with a different mean, either lower or higher (two-tailed test). *P*-values were determined with *n*-2 degrees of freedom. At a probability level of 0.1, only six samplers significantly came from a population with a mean not equal to one. Five of these were from a population with a mean normalized  $\alpha$  lower than one, while only one came from a population with a significantly higher mean  $\alpha$ . From this analysis it would appear that locations of rapid plume spreading are more transient than low-spreading sites.

To understand the spatial distribution of  $\alpha$  values throughout the study, fitted  $\alpha$  values were statistically tested to find whether the observations fit a normal or a log-normal distribution. Normalized  $\alpha$  values from each sampling event as well as the

pooled 78 dispersivities from all three tracer events were plotted in histograms. The distribution of all 78 normalized dispersivities revealed a distribution that appeared log-normal. Figure 7 shows the distribution of the log of the 78 normalized  $\alpha$  values, which on first inspection appears roughly bell shaped.

The Filliben test statistically compared the measured distribution to a theoretical normal distribution [7]. For this Filliben analysis, it was necessary to omit the value calculated by the number 17 sampler from the 1992 application. The  $\alpha$  value of this sampler was too small to include in the test for normality because the  $\text{Log}_{10}$  value of this near zero number had a very large absolute value. The Filliben test revealed that the  $\text{Log}_{10}$  values of the normalized dispersivities fit a normal distribution. The test was applied to each set of 26 values generated during each tracer application as well as the 78 pooled  $\alpha$  values.

### **3.5 Calculation of Transport Parameters Using Manual Soil Sampling**

Manual soil sampling in May and September 2001 provided a “snapshot” in time of the  $\text{Br}^-$  concentration in soil water throughout the upper 1.2 meters of the soil profile. The temporal transport parameters calculated by the PCAPS at a fixed depth through time were applied to the ADE to predict the  $\text{Br}^-$  profiles measured through soil sampling. Prediction of true soil  $\text{Br}^-$  concentration using temporal parameters could only be made on fallow plots due to  $\text{Br}^-$  redistribution on cover cropped plots.

Temporal parameters were not successful at predicting  $\text{Br}^-$  profiles at 143 days (May sampling) or 280 days (September sampling). Temporal parameters predicted a profile that was much more dispersed than the observed spatial profile (Figure 8). To satisfy the conservation of mass, peak  $\text{Br}^-$  concentrations were underestimated. The inability of the temporal parameters to predict tracer concentration at an adjacent location might either result from transport parameters being highly variable over space or hydrodynamic dispersion of the tracer plume is not constant in time. Due to temporal parameters consistently overestimating dispersion, the second explanation seems more appropriate.

Dry conditions with no intense rain events dominated the site preceding the soil sampling in summer 2001. This type of weather favors matrix flow and  $D_0$  spreading

processes. Temporal parameters calculated after May 2002 show the hydrodynamic dispersion of the dry 2001 water year in addition to the above average 2002 water year. Intense rainfalls favored high pore velocities and some preferential flow during the second year, causing the plume to spread and preventing transport parameters to be applicable to both halves (in time) of the tracer breakthrough. Effectively, it appeared that the ADE successfully fitted the concentration data measured from PCAPS, but was incapable of accounting for the transport mechanism dominating tracer transport throughout the breakthrough.

The ADE was fit to the spatial tracer profiles to generate transport parameters (spatial parameters). As suggested by Figure 8, spatially fitted dispersivities were much lower than the temporal parameters (Figure 9) in all but one location. Spatial dispersivities calculated in September were generally smaller than those calculated in May. This was due to the smaller pore water velocities calculated for September as very little additional water was collected from the PCAPS between May and September, mathematically lowering the average pore water velocity since tracer application. Four of the nine spatial profiles provided negative dispersivities in September. Such non-physical values were generated because of the method of calculating  $\alpha$  that accounted for  $D_0$  (Equation 11). The negative dispersivities reflect the fact that all spreading could be explained by  $D_0$  during dry summer months in the Willamette Valley.

#### **4. Conclusions**

Three applications of conservative tracers were made to vegetable plots over an eleven year period. Breakthroughs of the tracers at the 1.2 meter depth took place over several seasons and were predominately driven by winter rainfall. Preferential flow was observed at the site, but was not the dominant factor in tracer transport and spreading. Long periods of flow through the soil matrix resulted in unimodal tracer breakthroughs with steady rising and falling limbs. The ADE fit the data well.

Tracer breakthrough and transport parameters varied following each application. Dispersivity values calculated from different tracer events were found to have no correlation in time and a log-normal distribution in space. Variability of  $\alpha$  through time

could not be explained by pore water velocity. Dispersivity values calculated using PCAPS had no capacity to predict spatial  $\text{Br}^-$  concentrations for earlier times during the same breakthrough. Spatially calculated  $\alpha$  values during May and September 2001 could not capture the complex solute spreading processes occurring the following winter and expose the inability for one model parameter to describe solute spreading in natural conditions.

These results call into question the concept of a characteristic and static  $\alpha$  within soil systems; instead suggesting that differences in cycles between molecular diffusion and mechanical dispersion dominated periods were foremost in determining solute spreading. Dominant spreading processes were related to varying periods of preferential and matrix flow and the balance between more and less water mobile portions of the soil column. Transient flow along structural features between long periods of molecular diffusion dominated transport affected tracer spread in the system. These observations agree with those of Persson and Berndtsson [15] as they correlated smaller dispersivities with more consistent water application and less variation in water velocity.

These findings imply that the ADE has limited capability to predict solute spreading for structured soils under variable rainfall conditions. In view of the inability of the current conceptual model describing solute spread as dependent on a static  $\alpha$ , site specific parameters calculated from previous experiments should not be used until variations in water inputs, risk and room for error in the engineering application are considered.

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- 1 Monthly precipitation following each of the tracer applications. Monthly rainfall totals are given from November 1992 through October 1994, November 1995 through April 1997, and December 2000 through April 2003 for the 1992, 1995 and 2000 tracer applications, respectively. Vertical lines show points in time when integer numbers of pore volumes were collected by PCAPS.
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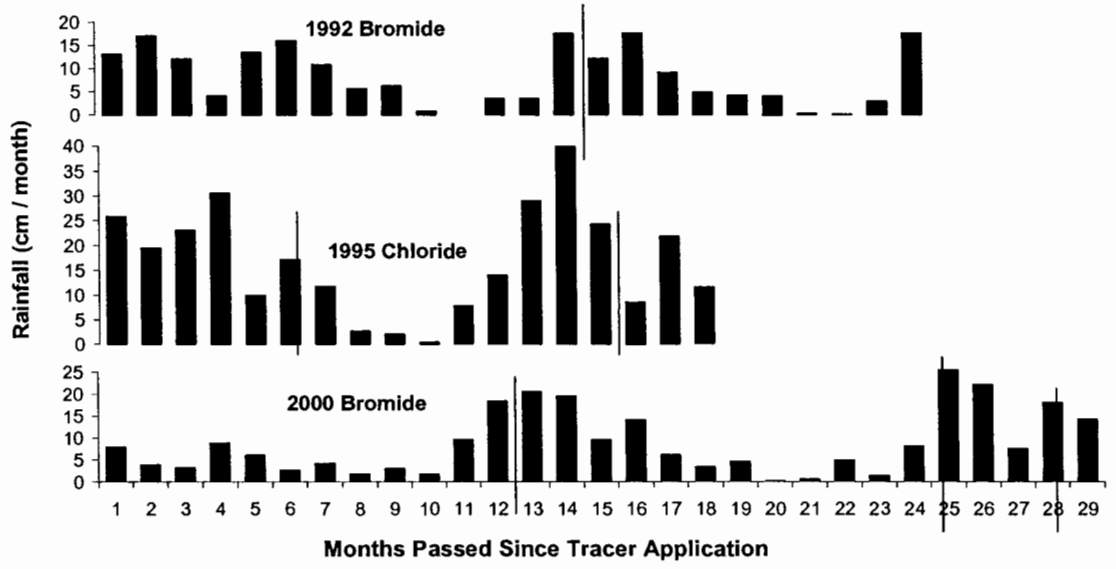


Figure 1

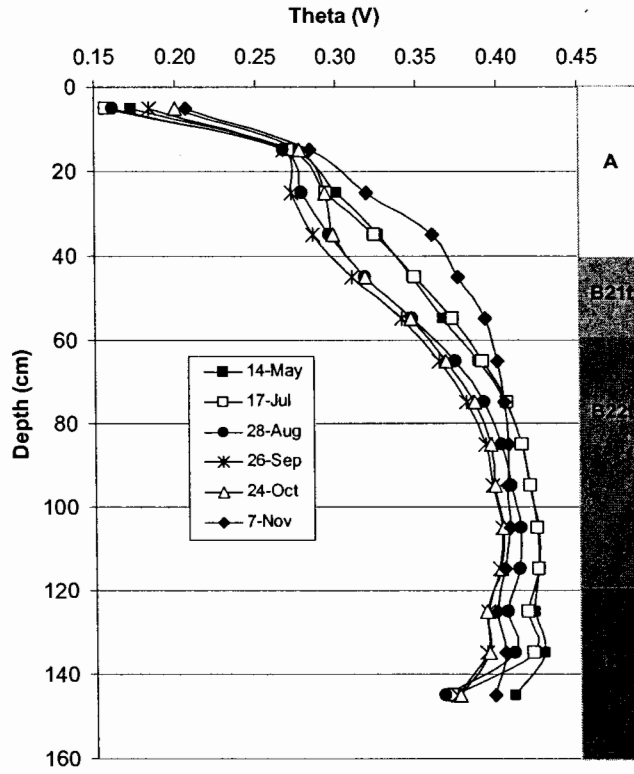


Figure 2

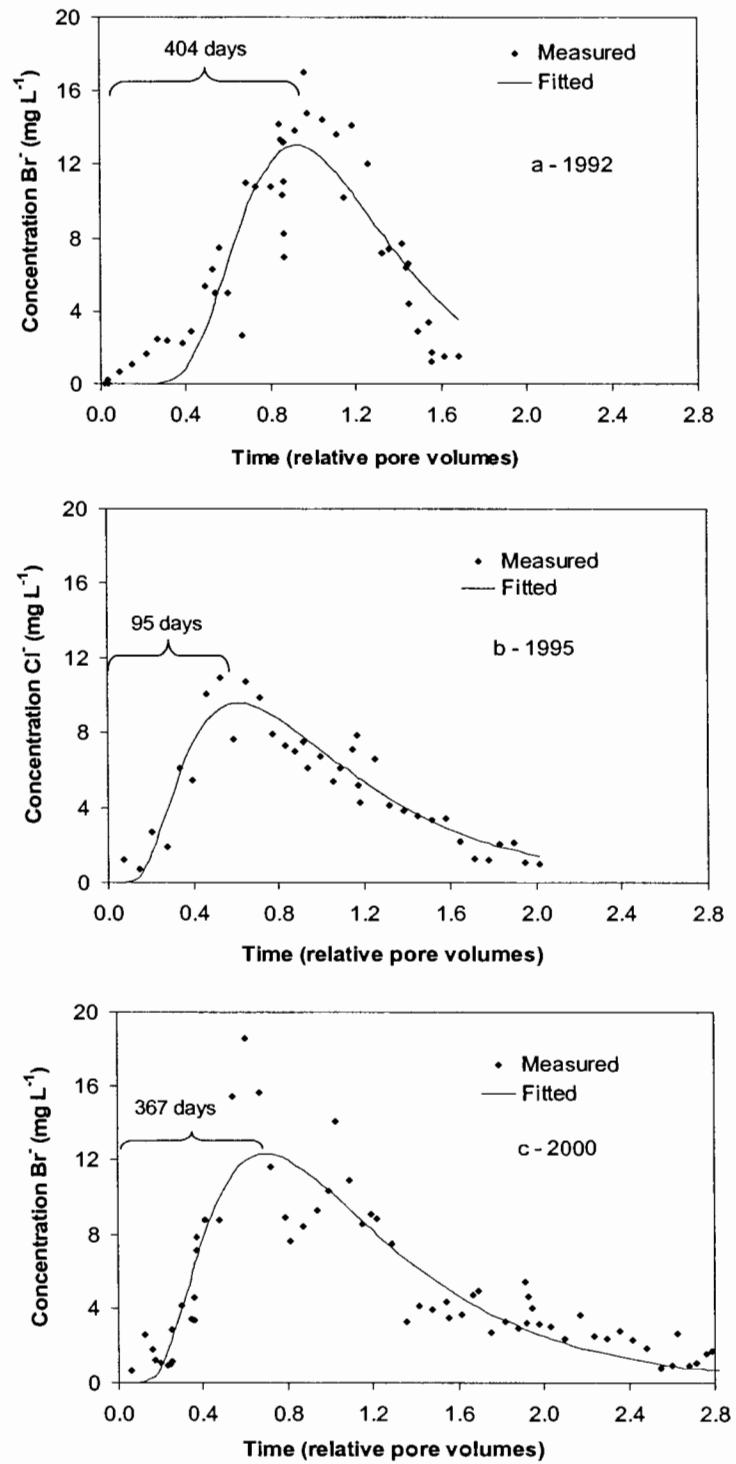


Figure 3

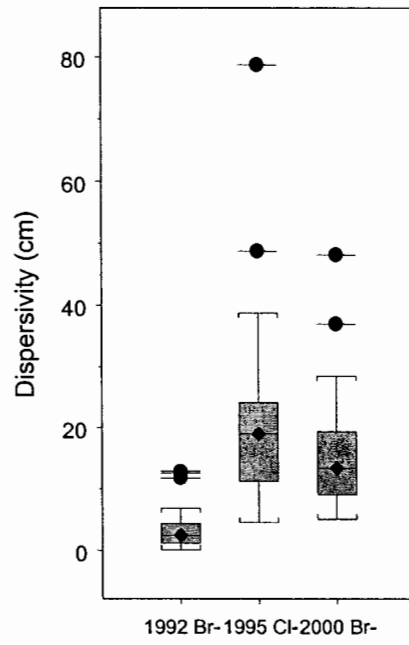


Figure 4

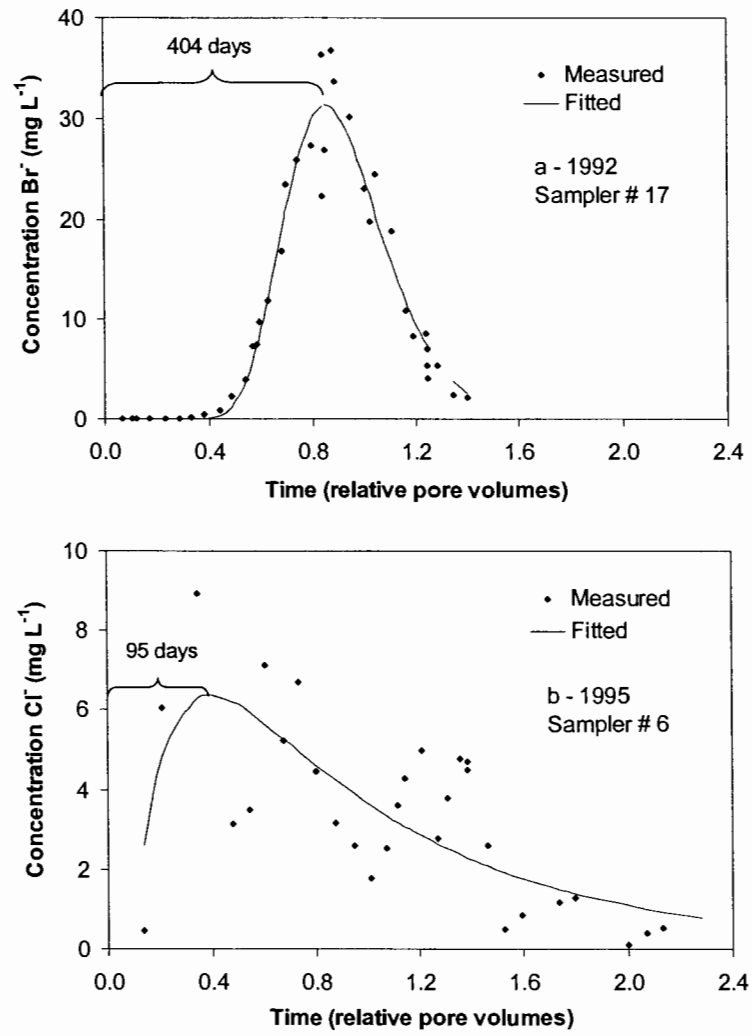


Figure 5

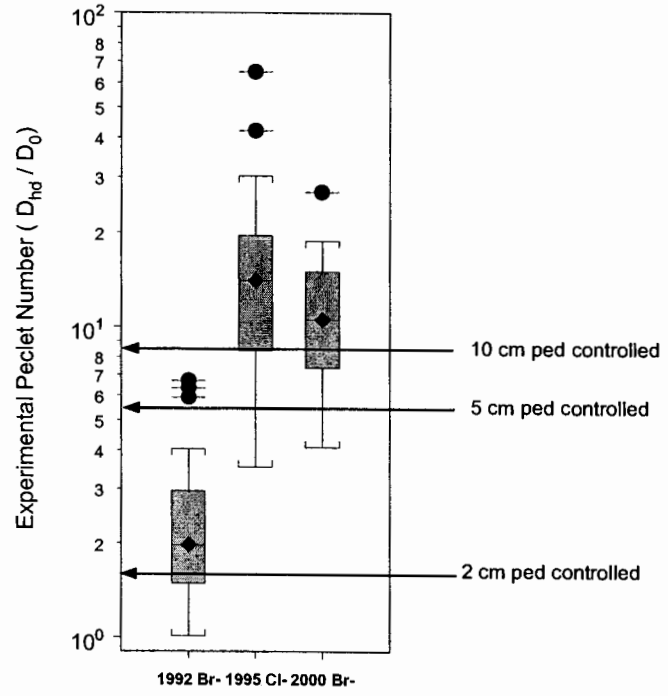


Figure 6

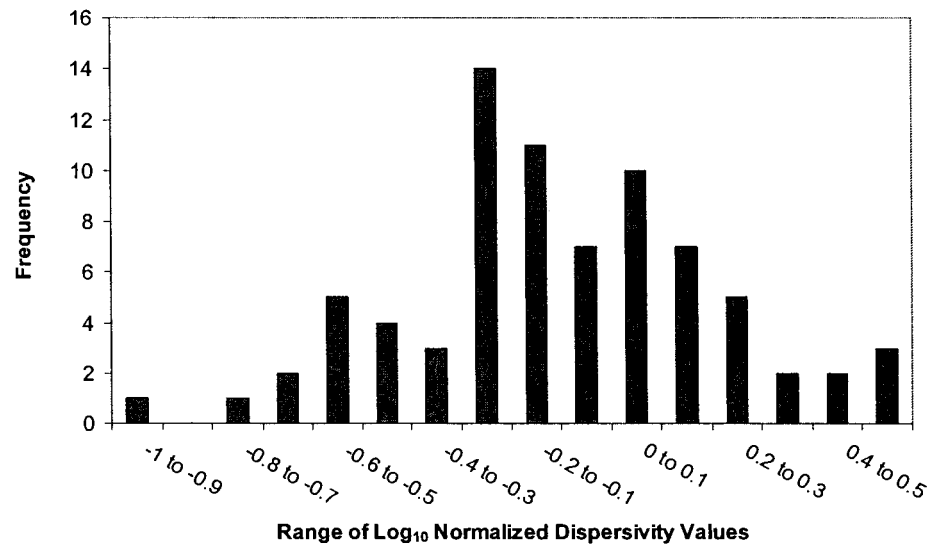


Figure 7

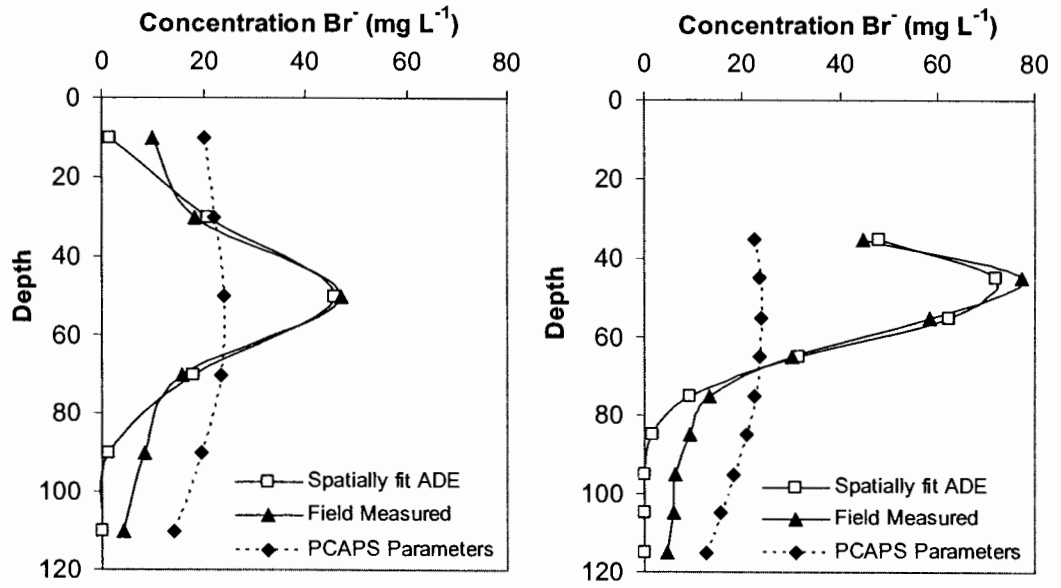


Figure 8

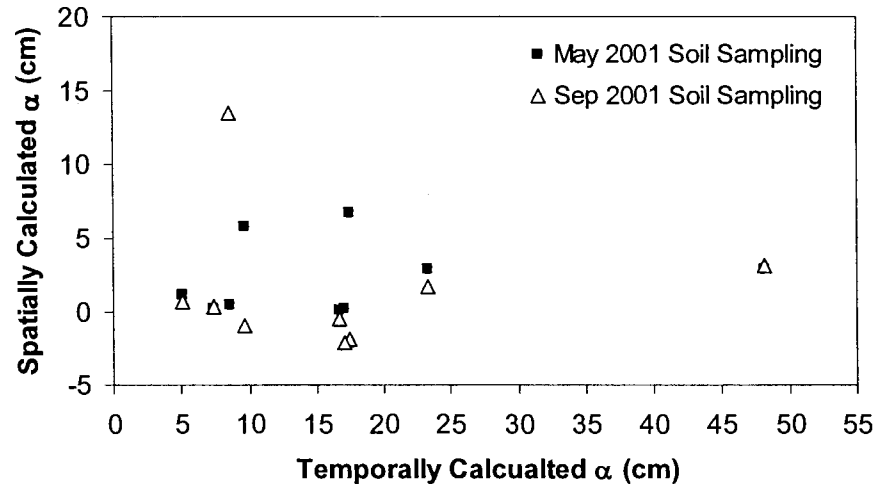


Figure 9

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- 2 Summary of analyses of variance showing the sources of effects on fitted  $\alpha$  values for all three applications grouped together and the 2000 Br<sup>-</sup> application alone. Cover crop treatment did not have a significant effect on  $\alpha$  though it did have a significant effect on bromide collection efficiency [6].

Table 1 Average and range of transport parameters for each conservative tracer application. Parameters fitted from tracer breakthrough measured through time by PCAPS lysimeters and spatially by soil sampling are shown.

<b>Summary of Transport Parameters</b>				
<b>Statistical Group</b>	<b><math>D_m</math> (cm<sup>2</sup>/h)</b>	<b><math>\alpha</math> (cm)</b>	<b><math>V</math> (cm h<sup>-1</sup>)</b>	<b>R</b>
1992 Br <sup>-</sup> Application	0.045 (2.29 x 10 <sup>-4</sup> , 0.164)	3.74 (0.024, 12.9)	0.012 (0.007, 0.014)	0.98 (0.60, 1.24)
1995 Cl <sup>-</sup> Application	0.455 (0.073, 1.84)	20.6 (4.48, 78.8)	0.022 (0.015, 0.026)	0.74 (0.23, 1.33)
2000 Br <sup>-</sup> Application	0.308 (0.089, 0.747)	16.1 (5.04, 48.1)	0.020 (0.011, 0.027)	0.84 (0.51, 1.32)
2000 Br <sup>-</sup> Application † May 2001 Soil Sampling	0.025 (0.002, 0.077)	2.29 (0.123, 6.79)	0.012 (0.006, 0.015)	0.75 (0.44, 0.99)
2000 Br <sup>-</sup> Application † May 2001 Soil Sampling	0.01 (-0.15, 0.109)	1.55 (-2.13, 13.5)	0.006 (0.003, 0.008)	0.79 (0.40, 1.17)

† Calculated only with holes augured from fallow plots.

Table 2 Summary of analyses of variance showing the sources of effects on fitted  $\alpha$  values for all three applications grouped together and the 2000 Br- application alone. Cover crop treatment did not have a significant effect on  $\alpha$  though it did have a significant effect on bromide collection efficiency [6].

Time Period	Source of Variation	Dispersivity ( $\alpha$ )	
		df	P-value
1992, 1995, and 2000 tracer applications	Year	2	< 0.0001
	Cover crop	1	0.5847
	N fert. rate	2	0.6551
	Block	3	0.2000
	Year * Cover crop	7	0.6312
	Cover * N fert. rate	2	0.3784
	Year * N fert. rate	14	0.3648
	2000 tracer application	Cover crop	1
	N fert. rate	2	0.7815
	Block	3	0.4587
	Cover * N fert. rate	2	0.8239