PNNL-13795

Vadose Zone Transport Field Study: Soil Water Content Distributions by Neutron Moderation

A. L. Ward
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G. W. Gee

October 2000

Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RL01830

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Pacific Northwest National Laboratory
Richland, Washington 99352

Summary

At Hanford, one of the principal uncertainties in vadose zone flow and transport are the current is caused by the natural heterogeneity of the soil in which the contaminants reside. The Vadose Zone Transport Field Study (VZTFS) was conceived as part of the U.S. Department Energy Groundwater/Vadose Zone Integration Project Science and Technology initiative to address the major uncertainties confronting vadose-zone fate and transport predictions at Hanford. During the last FY, a series of filed experiments was conducted with the goal of identifying the principal uncertainties limiting the interpretation of current contaminant distributions and prediction of future migration through the vadose zone.

This report summarizes the water-content data obtained during a series of five injections at the VZTFS Site during the summer of FY 2000. Water contents were measured by the neutron-moderation techniques in a series of 32 steel-cased boreholes extending down to a depth of about 17 m. Three-dimensional distributions of water content are used as a first step in trying to understand the mechanisms domination transport through Hanford's vadose zone.

A comparison of lithologic logs and neutron probe data show a continuous layer of higher θ, indicative of a fine-textured layer, between 5.5 and 7.0 m and sloping upwards in an easterly direction. A similar layer is also observed between 10.5 and 11.5 m. Because these fine-textured regions are underlain by coarser soils, they could very well act as regions of preferential lateral flow until the water-entry pressure of the coarser soils is exceeded. At this point, data analysis has been mostly qualitative with quantitative analysis planned for the first quarter of FY 01. Nevertheless, patterns in subsurface water-content distributions show a clear dependence on textural stratification, which suggests that conceptual models neglecting the occurrence of fine-textured lenses may grossly over predict the vertical migration of water and under predict the lateral migration of water and contaminants. Mass-balance calculations using neutron-probe data show relatively good agreement with the total amount of water added but not with individual injections.

Future activities include a more thorough analysis of the data, including the derivation of hydraulic properties by inverse procedures, and the evaluation of fluid properties on transport through the use of concentrated ionic tracer in early FY 01.

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1.0 Introduction

Contaminant transport through the vadose zone is a complex process controlled largely by interactions between subsurface lithologic features, water flow, and fluid properties. Understanding the processes controlling transport is an important prerequisite to the development and implementation of effective soil and groundwater remediation programs. However, difficulties in directly observing and sampling the subsurface can complicate attempts to better describe subsurface transport processes and is mostly responsible for the large amount of uncertainty associated with vadose-zone processes. The reduction of the uncertainty has been identified as a site need at Hanford by the STCG and the National Research Council and is a key aspect of the site's science and technology effort.

At Hanford, the principal uncertainties are the current distribution of source contaminants and the natural heterogeneity (over a range of spatial scales) of the soil in which the contaminants reside. Field-scale infiltration and tracer tests are well-established techniques for identifying vadose-zone flow and transport processes and characterizing uncertainty. Such tests have also been successfully used to determine the parameters needed to describe the relevant processes in interpretive and predictive models.

The Vadose Zone Transport Field Study (VZTFS) was conceived as part of the U.S. Department of Energy Groundwater/Vadose Zone Integration Project Science and Technology initiative to address the major uncertainties confronting vadose-zone fate and transport predictions at Hanford and to overcome the limitations highlighted above. The VZTFS is intended to conduct field investigations that will improve our understanding of field-scale transport and lead to the development or identification of efficient and cost-effective characterization methods.

2.0 Scope and Objectives

The scope and objectives of the VZTFS are described in detail by Ward and Gee (2000). Briefly, the scope is to conduct a series of field tracer experiments at Hanford to allow identification of the principal uncertainties limiting the interpretation of current contaminant distributions and prediction of future migration through the vadose zone.

The objective most relevant to this report is that aimed at identifying mechanisms controlling transport processes in soils typical of the hydrogeologic conditions of Hanford's waste disposal sites and to develop a detailed and accurate database of hydraulic and transport parameters for validation of three-dimensional numerical models. To achieve this objective, a series of experiments was designed to assure the measurement of flow and transport properties and their geostatistical properties in the same soil volume. The resulting data will form part of a data package to be used for calibrating numerical models and for calculating obtain field-scale transport parameters. They are intended to support the development of an upscaling methodology and the extrapolation of parameters derived from controlled field investigations at uncontaminated sites to contaminated sites with less characterization. The resulting information will be used to improve conceptual models and to direct the selection of remedial actions. These results will support the development of the vadose-zone component of the System Assessment Capability (SAC).

This report summarizes the water-content data obtained during a series of five injections at the VZTFS Site during the summer of FY 2000. Water contents were measured by the neutron moderation techniques in a series of 32 steel-cased boreholes extending down to a depth of about 17 m. Three-dimensional distributions of water content are used as a first step in trying to understand the mechanisms domination transport through Hanford's vadose zone. First, a brief review of the neutron moderation technique is presented.

3.0 Neutron Moderation Method

Subsurface neutron moisture gauges are widely used in hydrogeologic characterization of the vadose zone and particularly for estimating spatial and temporal distributions of soil water content, θ. The neutron technique is also used routinely to monitor field water contents at the Hanford Site (e.g., Ward and Gee 1997; Fayer et al. 1999, DOE 1999) and have been used extensively to monitor water content at the Sisson and Lu injection site (Sisson and Lu 1984; Fayer et al. 1993, 1995).

The instrument used in this study is the Campbell Pacific Nuclear (CPN) neutron hydroprobe (Model 503 DR), serial number H33115140. The probe includes a 50 mCi americium-241 and beryllium source and a neutron detector. The neutrons given off by the source (called "fast" neutrons) collide with the hydrogen atoms in soil water. Since the fast neutrons and the hydrogen atoms have the same mass, the fast neutrons are slowed down by this process, much like a billiard ball hitting a stationary ball of the same size and each moving away with equal speeds (one slowing down and the other speeding up). The detector measures only the slow neutrons; therefore, the amount of slow neutrons detected is directly related to the amount of hydrogen present. The main source of hydrogen is associated with water molecules and therefore soil water.

The radius of influence of the hydroprobe varies with source strength, hydrogen density (soil water content); particle density; and porous medium chemistry. Nevertheless, Olgaard suggested that the radius, r, accounting for 95% of the neutron flux in an infinite medium can be empirically described by

$$r = \frac{100cm}{1.4 + 10 \cdot ?_g} \tag{1}$$

where θ_g is the gravimetric water content in g cm^{-3}. Practical limits of the radius of influence in Hanford sediments may therefore range between 15 and 60 cm.

Using the neutron hydroprobe requires cased access tubes, 32 of which exist at the VZTFS Site. For measuring water contents, the probe was lowered in the vertical 15-cm i.d. steel-cased access tubes. Measurements were taken by first lowering the probe to the bottom of the well of interest after which counts were accumulated over a 15-s interval at 30-cm depth increments while withdrawing the probe from the well. Data were recorded by hand in the field notebook and electronically by the probe.

3.1 Neutron Probe Calibration

Neutron hydroprobes require calibration to convert counts for slow neutrons to soil volumetric water content. Fayer et al. (1995) provide a detailed description of the calibration method used by Sisson and Lu (1984) for the original experiment. Some questions have been raised about the actual calibration methodology. Nevertheless, a single calibration equation was developed for the three probes. In FY 2000, two probes (Serial No. H33115104 and H35056031) were used to log water content. Probe 1 (H33115104) was calibrated in the initial experiment and was used to cross-calibrate Probe 2 (H35056031) to assure continuity of the data. The probes were cross-calibrated by making measurements in the same well with the different probes. Cross-calibration measurements were made in Wells H2, H4, and 122.

The calibration function was developed by regressing water content, θ, on 15-s count times using a linear function. For Probe 1, Fayer et al. (1995) report a fitted function of the form

$$? = a + bC \quad (2)$$

where a and b are empirical constants equal to -6.98 and 0.0059, respectively. The coefficient of distribution, r^2, for Eq. 2 is 0.833. Probe 1 counts were regressed on Probe 2 counts also using a liner regression analysis. Figure 2 shows a plot of the observed data fitted to the relationship

$$?_{PRB.1} = a + bC_{PRB.2} \quad (3)$$

The regression coefficients are a = -8.677 and b = 1.5398 with a r^2 of 0.9903.

3.2 Monitoring Protocol

A baseline set of $\theta(x,z)$ was obtained on May 5, 2000. A series of five injections was made starting on June 1, 2000. During each injection, 4000 L of water were metered into a 5-m-deep injection well over a 6-hr period. Subsequent injections occurred weekly for a period of 5 weeks. The 32 steel-cased wells (Figure 1) surrounding the injection well were generally logged with a neutron probe within 24 hr of each injection except for the June, 26 injection. A wildfire approached within 0.5 km and access to the test site was restricted. The wells were logged on July 7 instead. Subsequently, two more readings of the 32 wells were completed during the month of July. An additional 4000-L injection was made on September 18 to obtain *in situ* hydrologic properties using a combination of pressure measurements and neutron-probe water-content measurements at the same depth.

Details of the experimental setup and suite of instruments used in the experiment are provided in the executive summary of the VZTFS (Ward and Gee 2000), which may be found at: http://etd.pnl.gov:2080/vadose/files/VZTFS-FY00Summary.pdf.

Both probes were used to monitor $\theta(x,z;t)$ during the experiment. Probe counts were accumulated over a 16-s interval. Counts from Probe 2 were converted to an equivalent 16-s count on Probe 1 using the cross-calibration relationship (Eq. 2). The spatial distribution of water content, $\theta(x,z)$ was determined using Eq. 1.

Discontinuous $\theta(x,z)$ obtained at the discrete sampling wells along the various transects were interpolated by kriging to produce continuous two-dimensional and three-dimensional distributions of $\theta(x,y,z)$ for the various times.

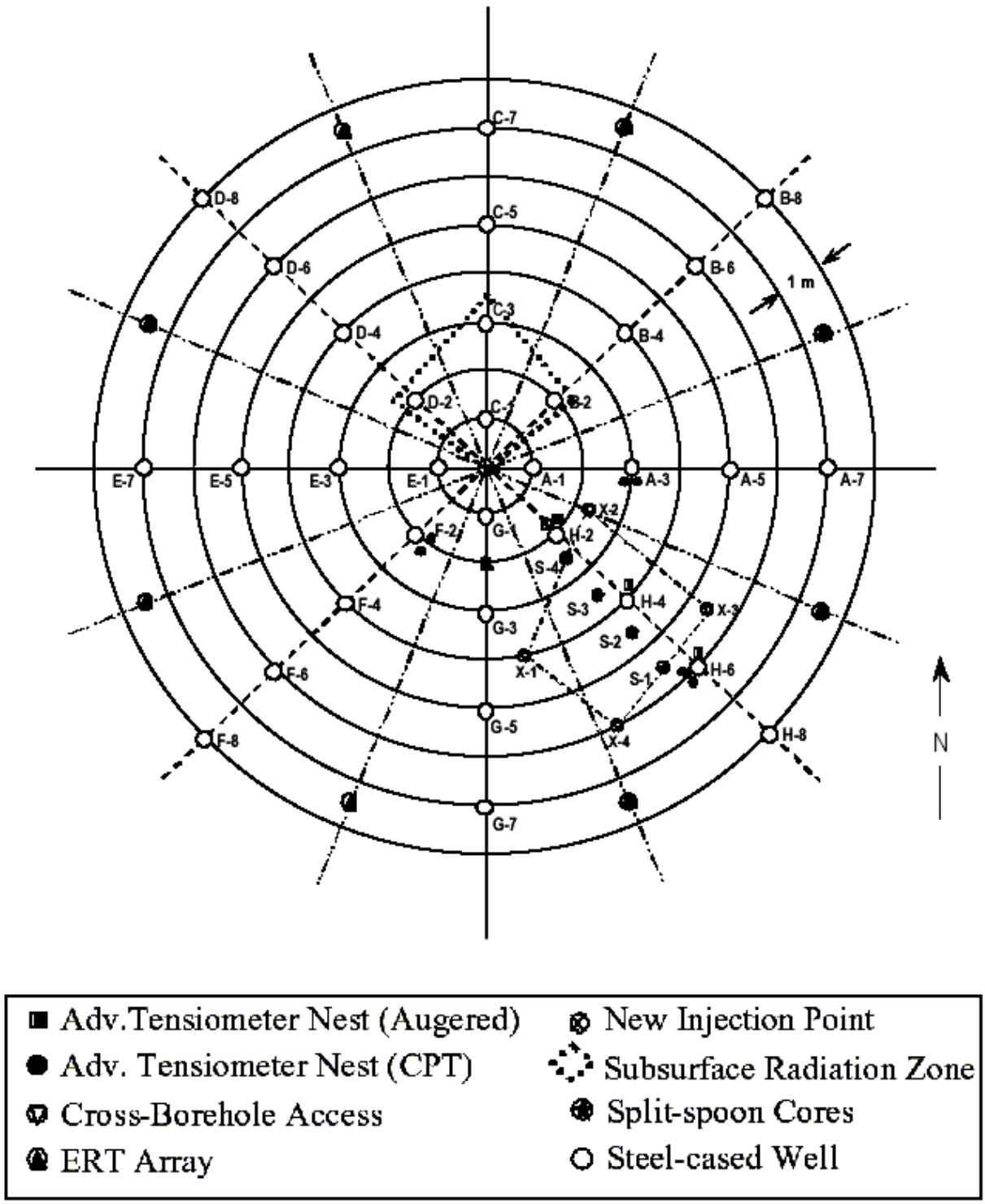

Figure 1. Plan View of FY 2000 Test Site Showing Locations of Access Tubes, Vertical Electrode Arrays, and Injection and Core-Sampling Points

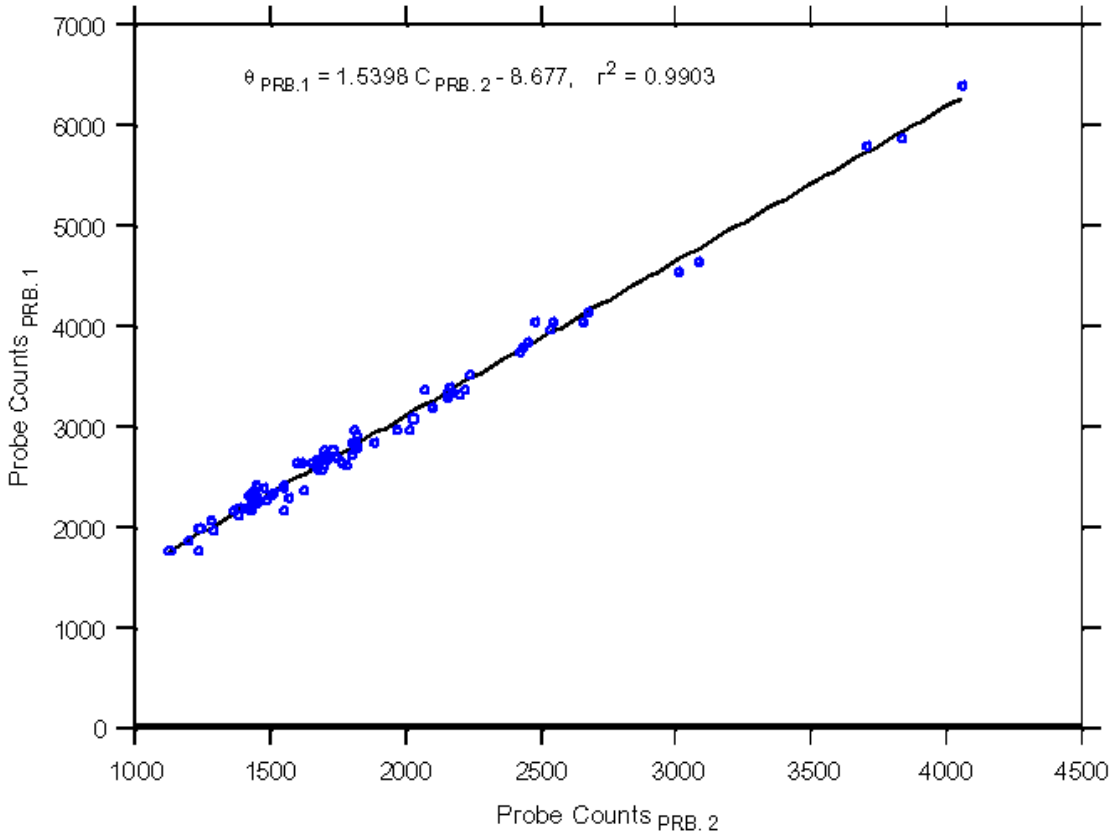

Figure 2. Linear Regression of 16-s Probe Counts with Probe 1 (H33115104) on Counts with Probe 2 (H35056031). Measurements made with Probe 2 were first converted to a Probe 1 equivalent with Eq. 2 following which θ was determined with Eq. 1.

4.0 Results

4.1 Identification of Major Stratigraphic Changes

Water-content distribution in soil profiles with steep gradients may be smoothed out with neutron-probe measurements, thereby reducing the level of detail. Nevertheless, neutron moisture logs have proven quite useful in identifying stratigraphic changes in the subsurface. Figure 2 shows examples of water content versus depth θ(z) profiles measured on May 5, 2000, in Well H4 and descriptive lithology logs obtained from adjacent cores S2 and S3 (Last and Caldwell 2001). These profiles represent essentially a steady-state condition as artificial injections of water were last made in September 1980, some 20 years before these measurements.

Neutron-moderation theory predicts relatively low count ratios, i.e., relatively low θ, in coarse-textured soils. Conversely, relatively higher count ratios and θ are observed in finer-textured soils such as fine sands, silts and clays. Thus, spikes in water content are generally coincident with silty fine-to-medium-fine sand. Figure 3 compares neutron logs of water content at Well H2 with lithologic logs obtained from soil cores taken adjacent to the well.

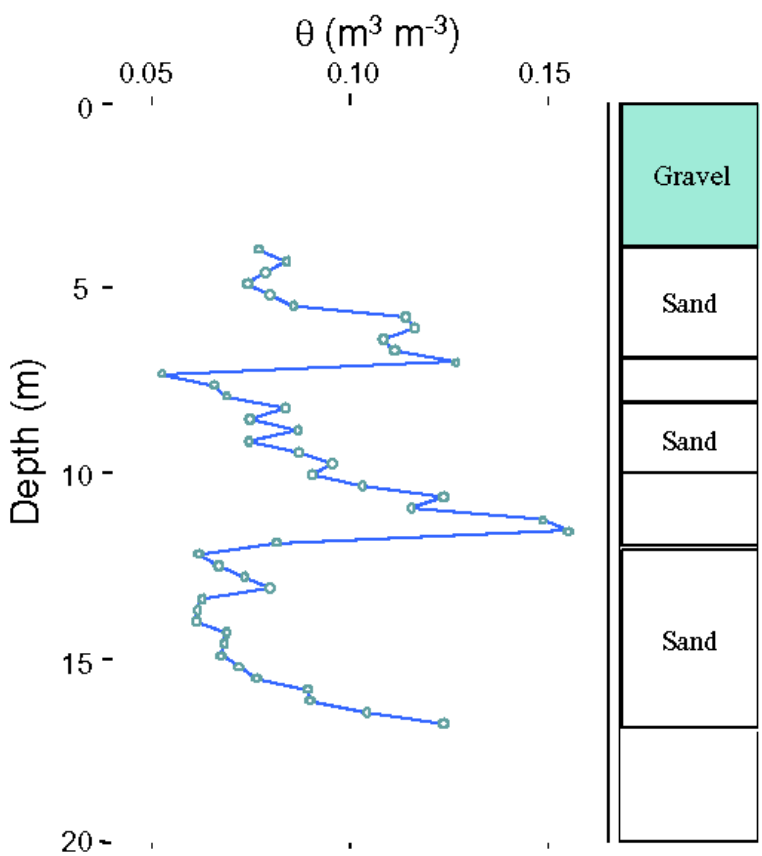

Figure 3. Neutron-Probe Measured Water Contents and Descriptive Logs for Well H2 at the VZTFS Site

Although lithologic logs were not obtained for the entire site, comparing the available logs and analyzing the more extensive neutron logs show a continuous layer of higher θ, indicative of a fine-textured layer, between 5.5 and 7.0 m and sloping upwards in an easterly direction. A similar layer is also observed between 10.5 and 11.5 m. Because these fine-textured regions are underlain by coarser soils, they could very well act as regions of preferential lateral flow until the water-entry pressure of the coarser soils is exceeded.

4.2 Spatial Distribution of Soil Water

Neutron-probe counts from the 32 wells were converted to θ(x,z) for each transect using Eq. 2 and Eq. 3. Data were interpolated using an anisotropic kriging algorithm in the Surfer software package. Figures 4 through 11 show two-dimensional contour plots of the resulting θ(x,z) distributions.

Figure 4 shows data collected on May 5, 2000, before any water injections and some 20 years after the tests conducted by Sisson and Lu (1984). The distribution of q shows a relatively uniform pattern except for two distinct regions of higher water content. The location of these regions has been correlated with the occurrence of soils with a texture finer than the main matrix. The first layer occurs between 5.5 and 7.0 m and slopes gently upwards in an easterly direction. A second fine-textured layer occurs between 10.5 and 11.5 m and shows very little slope. These layers could play an important role in infiltration and the subsequent distribution of water and contaminants.

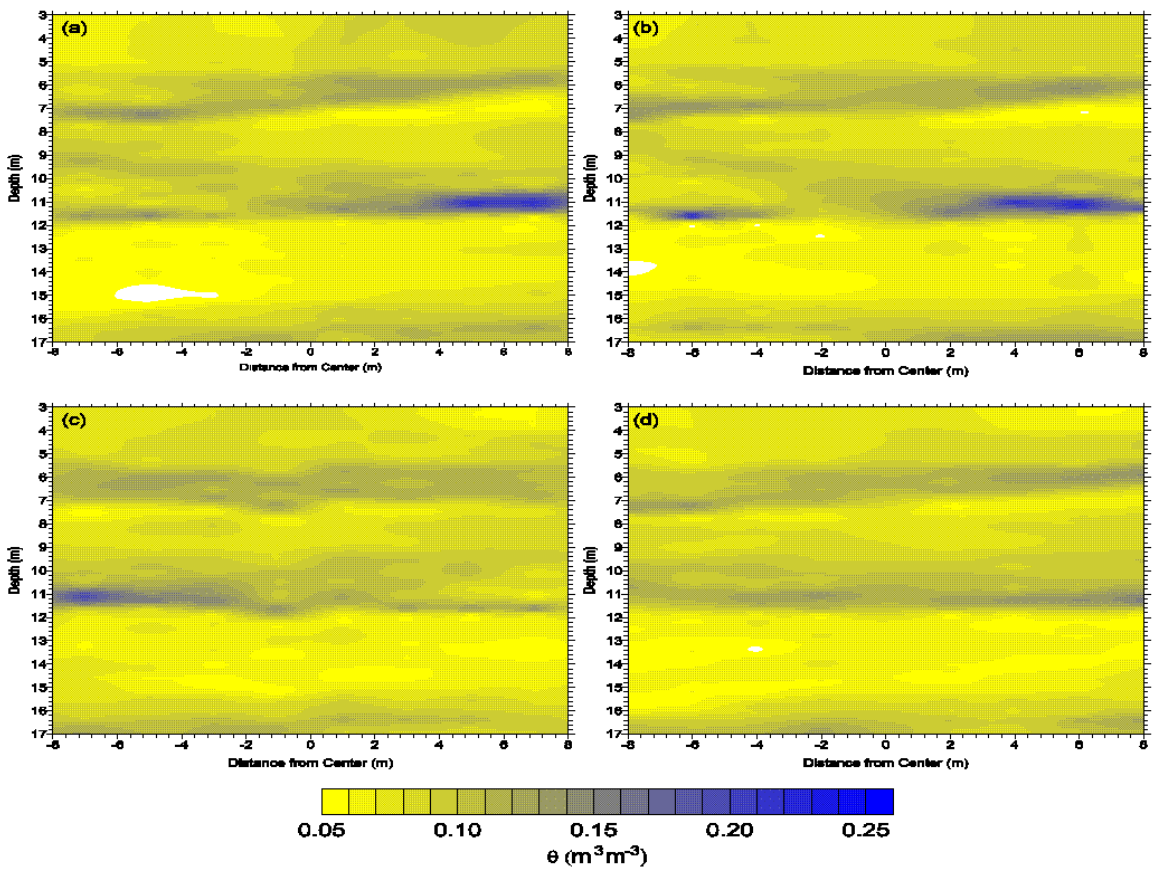

Figure 4. Spatial Distribution of Soil Water Content, θ, Interpolated from Neutron Probe Measurements in the 32 wells at the Vadose Zone Test Facility Shown in Figure 1 on May 5, 2000, Before any Water Injections: (a) Transect E-A, (b) Transect D-H, (c) Transect C-G, and (d) Transect B-F.

Figure 5 shows a similar plot using data collected on June 2, 2000, one day after the first injection of 4000 L of water. The injection point is located 5 m below the surface and about 1 m east of the center (x = 0 m) on the figure. This figure shows a somewhat nonuniform distribution of water beneath the injection point. In homogenous media, water infiltrating from a buried point source tends to form a symmetrical wetting bulb that becomes more elongated in the vertical direction as the texture of the soil becomes coarser. Here we see a wetted region that expands laterally, mostly in the region of the finer textured material at 5 to 6 m. Most of the increase in θ occurs in the D-H transect, suggesting that the water is flowing preferentially in a southeasterly direction. The fine-textured soil overlaying a coarse material is behaving like a capillary barrier and impeding the vertical migration of the water. Unlike in the other three transects, there is also a secondary wetting bulb developing beneath the main region along D-H (Figure 5b). This suggests that the matric potential is already high enough to overcome the water-entry pressure of the underlying layer, leading to breakthrough into the coarser layer.

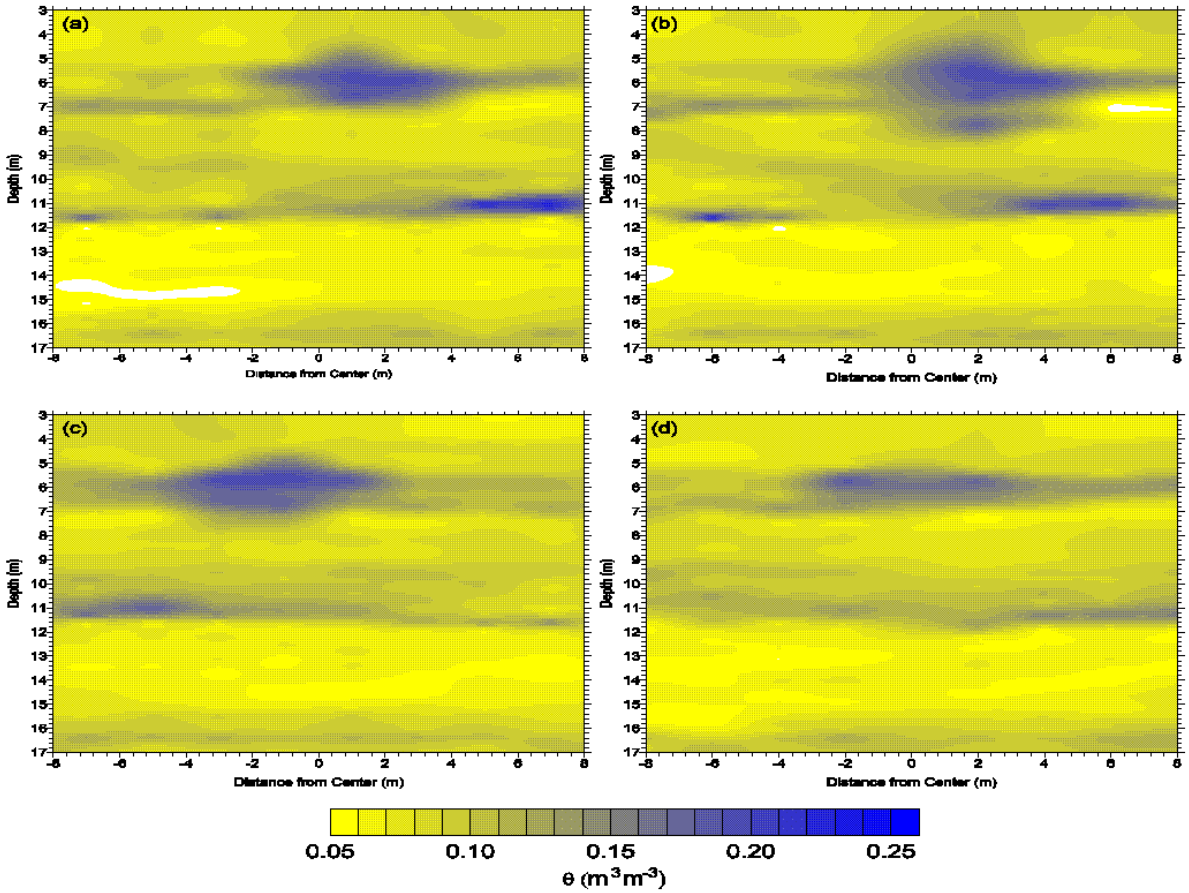

Figure 5. Spatial Distribution of Soil Water Content, θ, Interpolated from Neutron Probe Measurements in the 32 Wells at Vadose Zone Test Facility on June 2, 2000, One Day After an Injection of 4000 L of Water: (a) Transect E-A, (b) Transect D-H, (c) Transect C-G, and (d) Transect B-F

This observation could have implications on our interpretation of the behavior of subsurface leaks. Layering can serve to reduce the vertical migration of water and contaminants. Nevertheless, preferential movement in layered soils could result in the funneling of water and localized deep penetration once the water-entry pressure of the confining layer is exceeded.

Given that the θ_{max} observed in this study was generally less than 0.25 m^3 m^{-3}, this mechanism could become important with relatively small volumes of water.

Figure 6 shows a similar plot of data collected on June 9, 2000, again one day after a 4,000 L injection. By this time, enough water has been added to the system to cause penetration into the underlying coarser layer on the E-A transect (Figure 6a) and the C-G transect (Figure 6c). Along the D-H transect, the wetting front continues to expand beneath the fine layer and is beginning to impact the region immediately above the deeper fine layer (Figure 6b) while the B-F transect shows little change. It is clear that transport is primarily to the southeast.

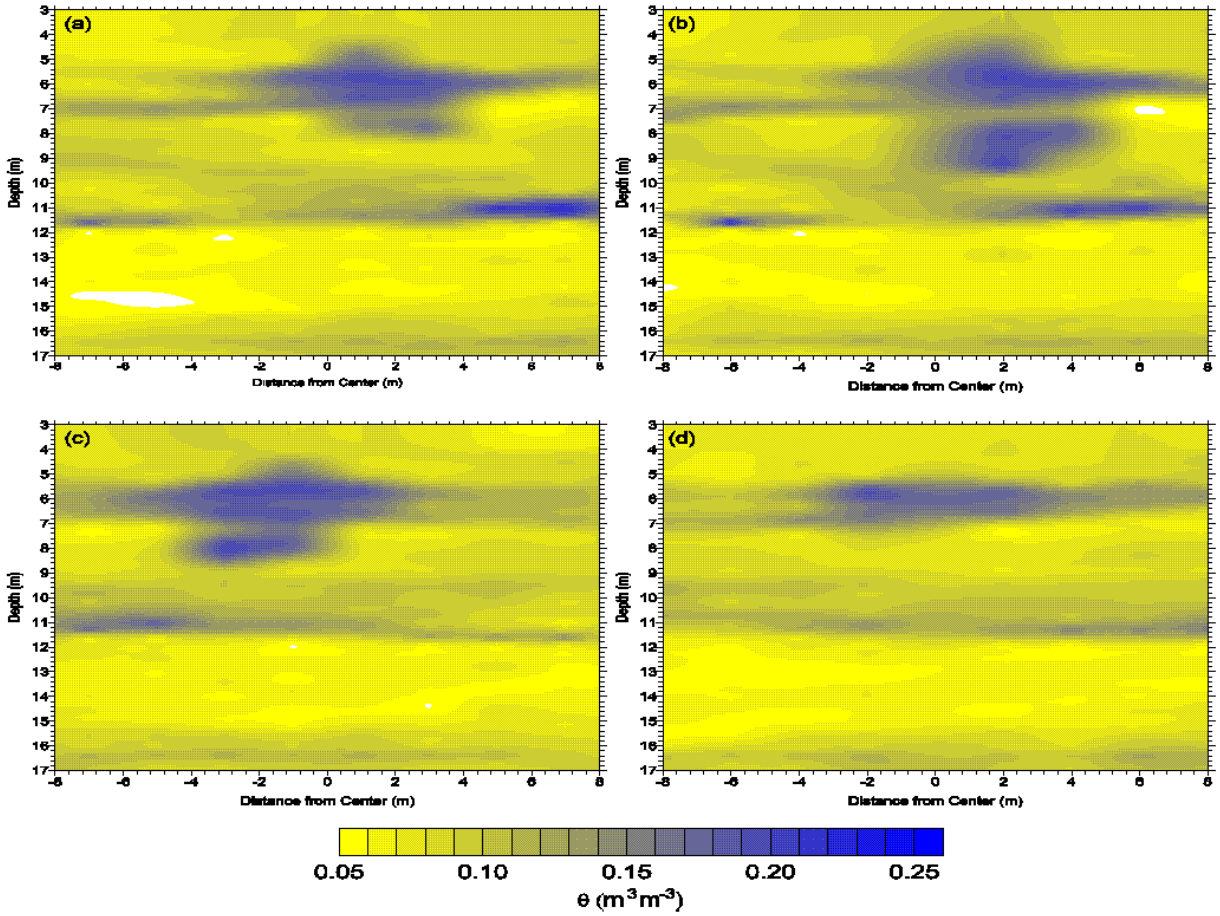

Figure 6. Spatial Distribution of Soil Water Content, θ, Interpolated from Neutron Probe Measurements in the 32 Wells at Vadose Zone Test Facility on June 9, 2000, One Day After an Injection of 4000 L of Water: (a) Transect E-A, (b) Transect D-H, (c) Transect C-G, and (d) Transect B-F

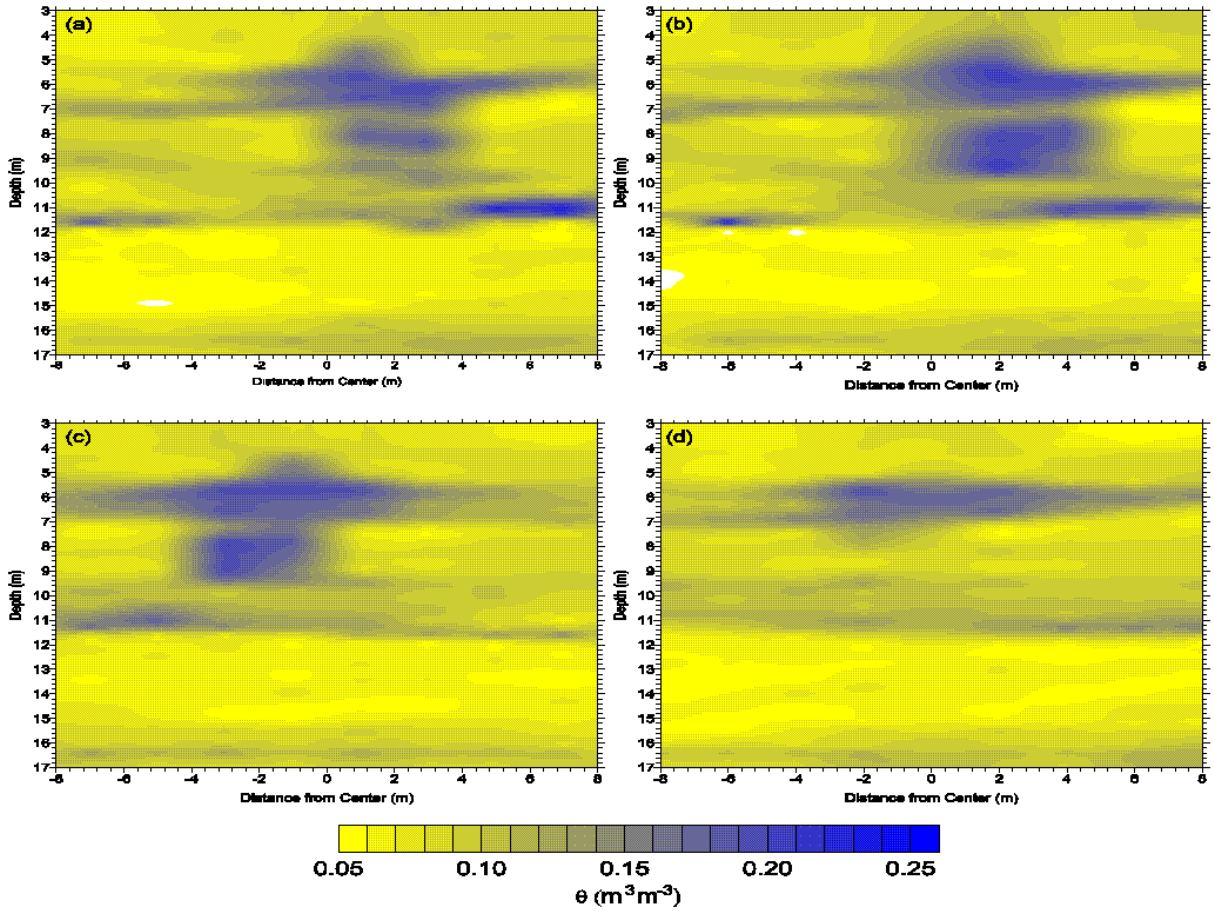

Figure 7. Spatial Distribution of Soil Water Content, θ, Interpolated from Neutron Probe Measurements in the 32 Wells at Vadose Zone Test Facility on June 16, 2000, One Day After an Injection of 4000 L of Water: (a) Transect E-A, (b) Transect D-H, (c) Transect C-G, and (d) Transect B-F

The remaining plots clearly demonstrate the anisotropic nature of the subsurface at this site and its effects on the movement of water. By June 23, 2000 (Figure 8), water penetrated down to the second fine-textured layer. However, water does not penetrate this layer, an observation that is confirmed by analysis of water samples for deuterium concentrations. Another interesting observation is that by this time, water was beginning to move out of the monitoring volume at the 5 to 6 m depth and the FB-F transect was also beginning to show an increase in water content. Data from July 7, 2000, only 9 days after the last injection, shows advanced redistribution. Although there is a significant buildup in water content near edge of the D-H transect, there is still no evidence of water penetrating the impeding layer at the 12-m depth. The remainder of measurements in July show a continuation in redistribution with the plume becoming discontinuous, particularly in the near-surface regions. By July 31 (Figure 11), after the injection of 20,000 L of water, most of the water had migrated outside of the sampling region and θ(x,y,z) was almost indistinguishable from the initial condition on May 5, 2000.

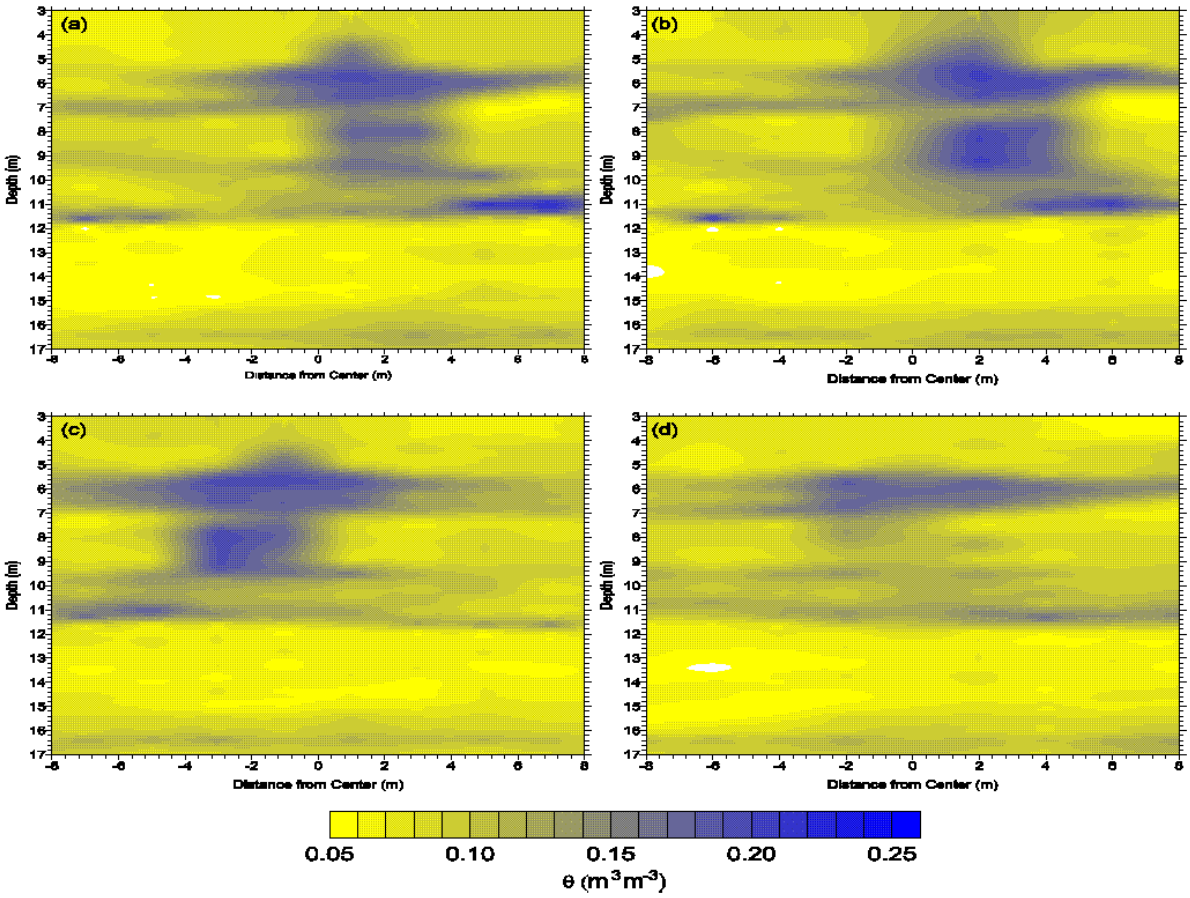

Figure 8. Spatial Distribution of Soil Water Content, θ, Interpolated from Neutron Probe Measurements in the 32 Wells at Vadose Zone Test Facility on June 23, 2000, One Day After an Injection of 4000 L of Water: (a) Transect E-A, (b) Transect D-H, (c) Transect C-G, and (d) Transect B-F

To this point, the analysis has been mostly qualitative with quantitative analysis planned for the first quarter of FY 01. Nevertheless, it is clear that conceptual models that neglect the occurrence of fine-textured lenses may grossly over predict the vertical migration of water and under predict the lateral migration of water and contaminants. It is possible that fluid behavior in the subsurface may be influenced by fluid properties such as density, surface tension, and viscosity. These factors are currently being studied in a series of laboratory experiments. These factors will be studied at the field scale in the next series of VZTF experiments in FY 01.

Data from the two-dimensional transects were combined to generate full three-dimensional plots of water content, which were then analyzed to estimate mass balance for the experiments.

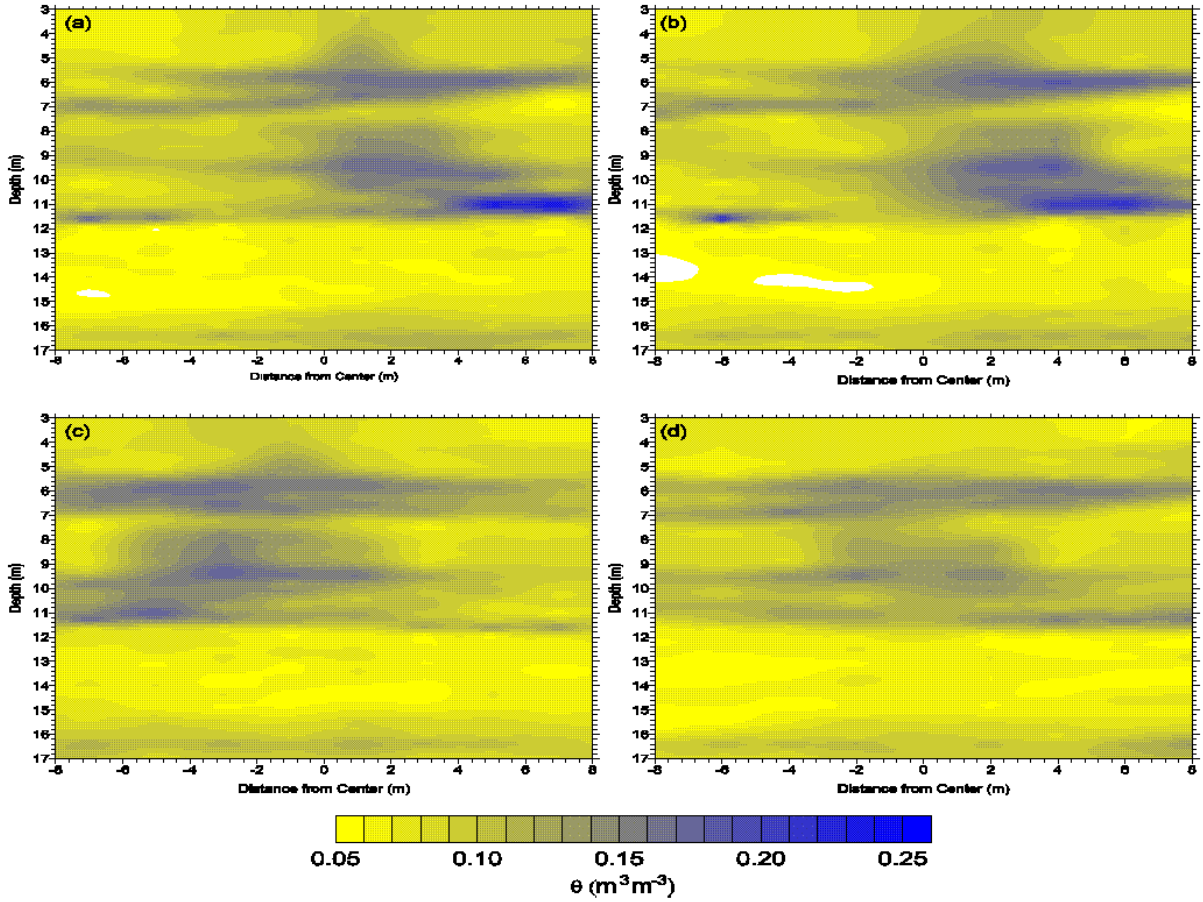

Figure 9. Spatial Distribution of Soil Water Content, θ, Interpolated from Neutron Probe Measurements in the 32 Wells at Vadose Zone Test Facility on July 07, 2000: (a) Transect E-A, (b) Transect D-H, (c) Transect C-G, and (d) Transect B-F

4.3 Mass Balance

To evaluate the mass balance in the system following the water injections, the volume of water in the monitoring domain was calculated for each set of neutron-probe measurements using an iterated integral. For a single-valued continuous function θ(x,y,z), in a closed region R, the volume of R, VR is given by

$$V_R = \iiint_R f(x, y, z,) dz\, dy\, dx \tag{5}$$

The spatial distribution of θ(x,y,z) measured by neuron probe, while singled valued, is not continuous, and Eq (3) is estimated as

$$V_R = \sum\sum\sum ?(x, y, z)?\, z\, ?\, y\, ?\, x \tag{6}$$

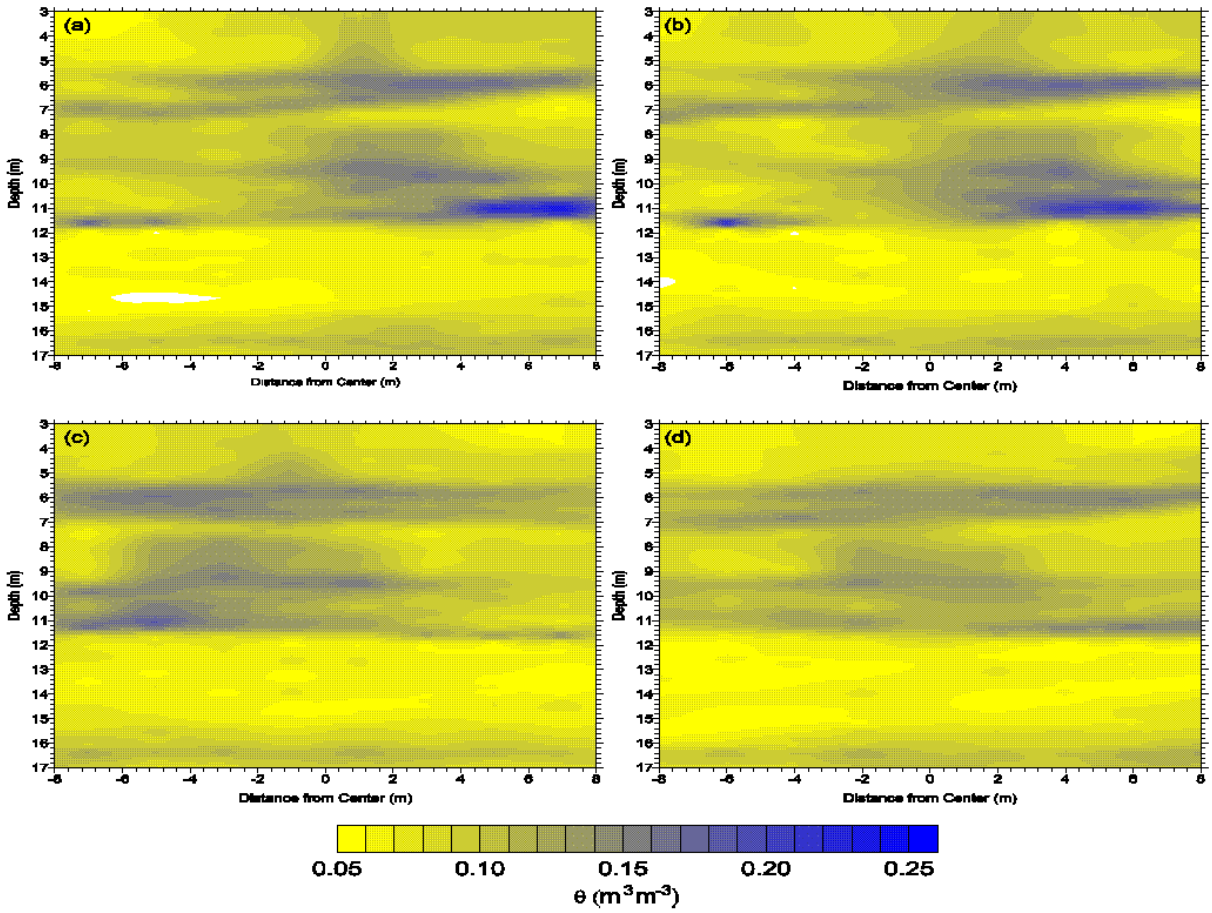

Figure 10. Spatial Distribution of Soil Water Content, θ, Interpolated from Neutron Probe Measurements in the 32 Wells at Vadose Zone Test Facility on July 17, 2000: (a) Transect E-A, (b) Transect D-H, (c) Transect C-G, and (d) Transect B-F

To evaluate Eq. (4), Measured θ(x,y,z) were interpolated between measurement locations using an anisotropic kriging algorithm with a fixed Δx, Δy, and Δz. Figures 12 and 13 show three-dimensional distributions of θ resulting from such a procedure using data described above.

Each figure is made up of 140,608 voxels, each with a unique value of θ. The volume of water in the sampling domain was calculated as the product of the volume of a single voxel (0.02 m^3) and the sum of θ over all of voxels. Table 1 shows the results of the mass-balance calculations for each neutron probe measurement. Recall that five injections, each of 4,000 L, were made during the course of the experiment for a total of 20,000 L.

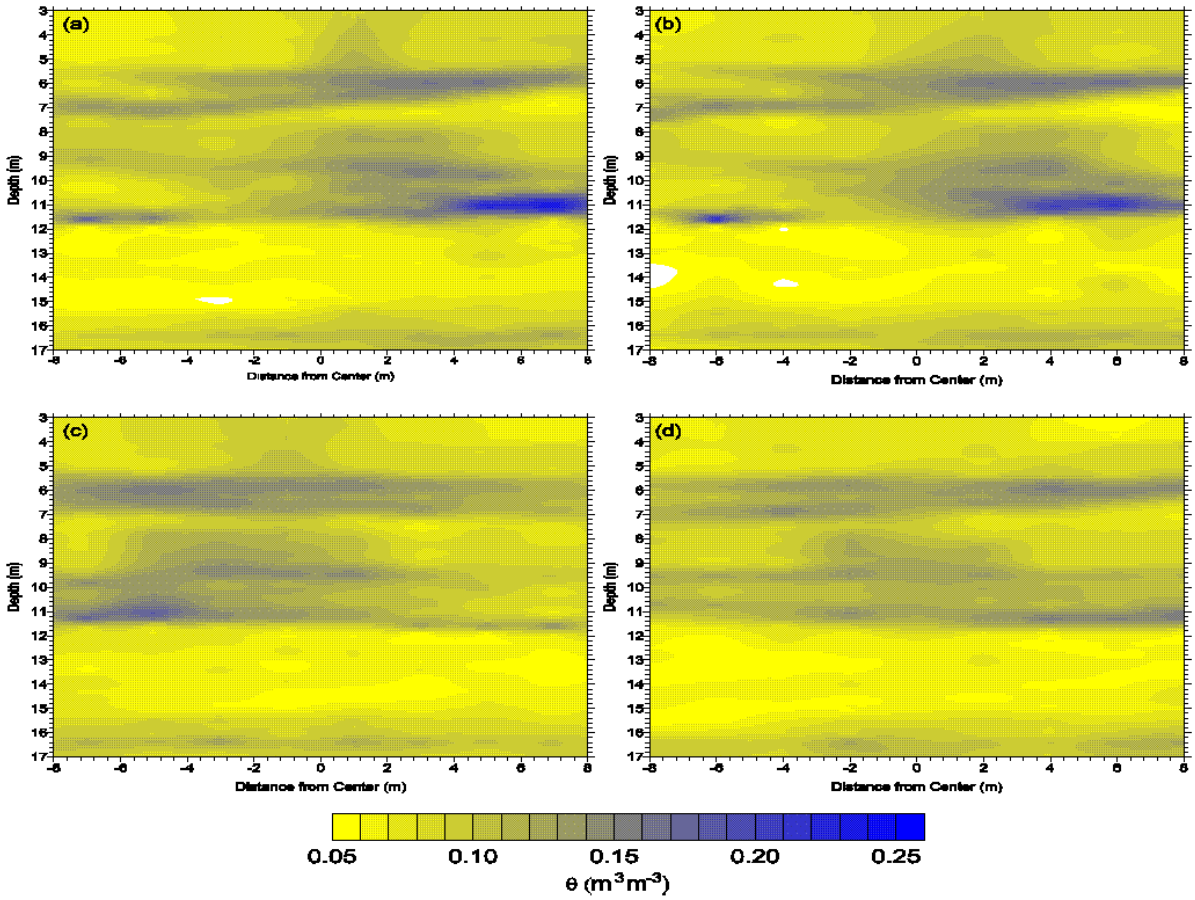

Figure 11. Spatial Distribution of Soil Water Content, θ, Interpolated from Neutron Probe Measurements in the 32 Wells at Vadose Zone Test Facility on July 31, 2000: (a) Transect E-A, (b) Transect D-H, (c) Transect C-G, and (d) Transect B-F

Table 1 shows that by the end of the injections (Injection No. 5 on June 28, 2000), a total of 19.37 m^3 of water had been injected compared the actual total of 20 m^3. However, a closer look at the mass recovery from individual injections shows some discrepancy between the observed and calculated volumes. This could be due to a variety of reasons. First, water was metered using a pre-calibrated flow meter with a planned application of 666 L per hr for a total of 6 hr. During the first injection, problems with the injection system resulted in flux almost twice the desired rate and the flow meter going off scale. It is possible that much more than 4 m^3 were injected during the first test, but 6.4 m^3 seems somewhat high. Subsequent tests were much better controlled; however, mass recovery still shows some discrepancy for the desired additions. This leads us to another possible explanation.

Although the basis for our analysis is the assumption of a continuous function θ(x,y,z), data collected with the neutron probe are not continuous. As discussed above, the radius of influence of the neutron probe varies with θ and with wells spaced 2 or more meters apart; there would have been several regions outside the range of the probe, especially at the higher water contents.

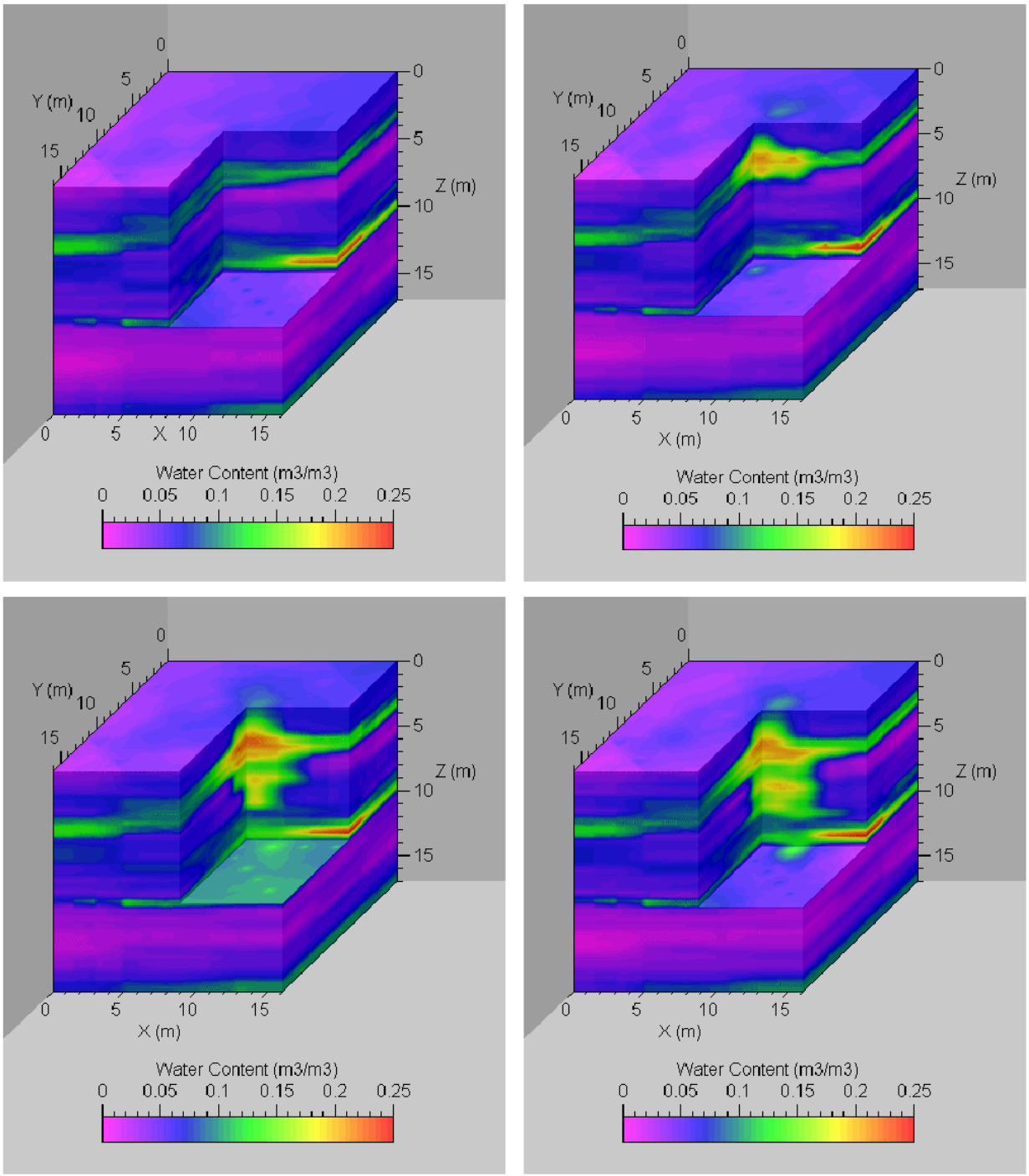

Figure 12. Spatial Distribution of Soil Water at the VZTF Site. Measurements were made on (a) May 5; (b) June 2; (c) June 9; and (d) June 16. Each three-dimensional plot is made up of 140,608 voxels with Δx =0.28 m, Δy = 0.28 m, and Δz=0.25 m.

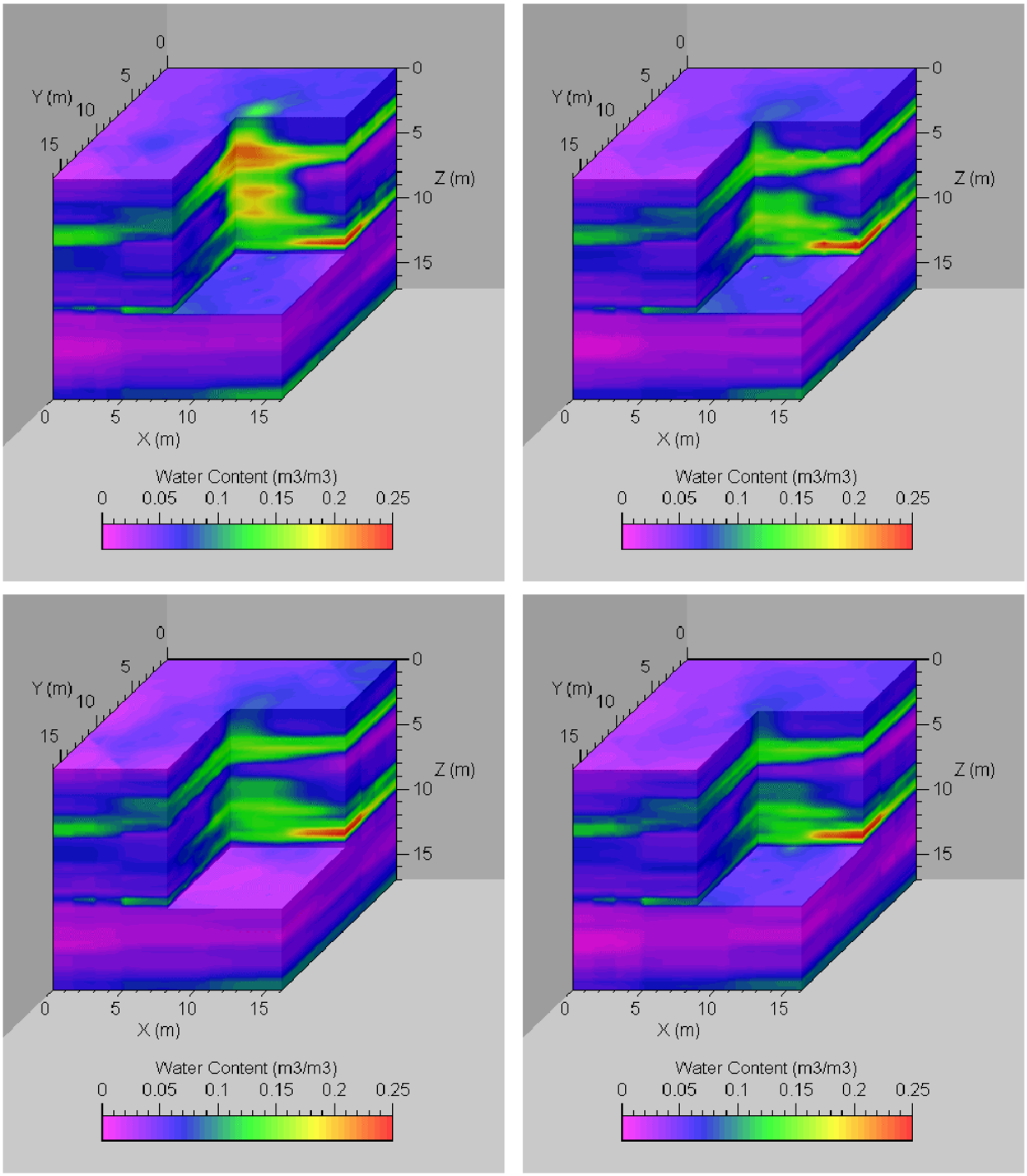

Figure 13. Spatial Distribution of Soil Water at the VZTF Site. Measurements were made on (a) June 23; (b) July 7; (c) June 17; and (d) July 31. Each three-dimensional plot is made up of 140,608 voxels with Δx =0.28 m, Δy = 0.28 m, and Δz=0.25 m.

Table 1. Mass Balance Calculations for the Five Water Injections Made During the Vadose Test Transport Field Test in FY 2000. Each injection consisted of 4 m^3 (4000 L) of water.

Sample No.	Sampling Date	$\sum\sum\sum \theta$ (x,y,z) (m^3 m^{-3})	V_R (m^3)	$V_{Ri} - V_{R1}$ (m^3)	ΔV_R (m^3)
1	5-May	11886.03	238.91	0.00	0.00
2	2-Jun	12206.00	245.34	6.43	6.43
3	9-Jun	12431.24	249.87	10.96	4.53
4	16-Jun	12687.75	255.02	16.11	5.16
5	23-Jun	12873.68	258.76	19.85	3.73
6	7-Jul	12849.60	258.28	19.37	-0.48
7	17-Jul	12764.39	256.56	17.65	-1.71
8	31-Jul	12686.52	255.00	16.09	-1.56

Furthermore, a closer look at the layout of the test site shows that the data were irregularly sampled. Irregularly sampled data are somewhat more complicated than those of regularly spaced data sets, and a viable strategy for analysis may involve resampling a given series onto a regular grid before conventional tools can be used for analysis. Resampling requires interpolation, and although there are several schemes available for this preliminary analysis, a simple kriging procedure was used. This approach may well have caused an overestimation of θ at some locations, leading to an overestimation of the recovery. This could be further compounded by the calibration coefficients, which according to Fayer et al. (1995) were questionable for the original 1980 experiments. During the first quarter of FY 01, these data will be more thoroughly analyzed and the calibration coefficients for the neutron probe and flow meter revisited.

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