Project Description

Context of Proposed Study

At the US Department of Energy Hanford Site a $^{43}\text{Tc}_{99}$ spill has been migrating horizontally through the vadose zone within a layer with enhanced fine components, although it was initially expected that the Tc would be transported vertically to the water table. This result is interpreted to imply anisotropy in $K$ at low moisture contents with horizontal values, $K_h$, larger than the vertical, $K_v$. Such anisotropy is attributed specifically to layers with higher silt content, relating properties on the scale of 10’s of meters to pore-scale characteristics. The anisotropy is thus believed to be traceable to higher $K$ values at high tensions (dry conditions) in the sediments with the greater fine component. This interpretation, although also proposed independently by PNNL researchers (e.g., Dr. Mark Rockhold), may not be valid. Comparison of the data for the suite of available soils considered relevant to the Tc spill with previous Hanford site data (Rockhold et al., 1988) shows that a number of these soil samples may well have lower conductivities than expected, implying that the interpretation that the delay in vertical transport is due to anisotropy at low moisture contents may be incorrect. An alternative possibility is that the anisotropic conditions apply at higher moisture contents but that at low moisture contents $K$ values are so low that Tc transport essentially does not occur. In order to predict future behavior of the spill it is necessary to distinguish at least between these two possibilities.

DOE grant DE-FG02-05ER64067 supported initial study of this problem. The initial research had two principle tasks. 1) to extend the existing treatment of anisotropy using the Rieu and Sposito (1991) fractal model and percolation theory to predict reliably distributions of hydraulic conductivity values, 2) to construct a model analogous to the Rieu and Sposito (1991) pore-scale truncated random fractal model allowing adoption of the results of 1 to the parameters of 2 and predictions relevant for the Tc spill. The first part could be done entirely at Wright State University, but the latter required direct interaction with PNNL scientists. Although part 1 of the research was expected to be finished before the beginning of funding, numerical calculations have only recently (beginning of November) delivered the result hoped for. The paper summarizing this research has now been submitted for publication. In regard to part 2, PI Allen Hunt spent two weeks in the summer of 2005 visiting PNNL. Chief contact was Dr. Glendon Gee (now retired) with some additional interaction with Dr. Mark Rockhold. In this time I proposed to the Hydrology group the novel method of studying the Hanford vadose zone based on application of percolation theory to a binary model of the subsurface, and collected several data sets. The binary model consisted of treating all the samples as sands, but distinguishing two kinds as either containing or lacking finer components. The samples with fine components were presumed to correspond to regions with higher moisture contents and higher hydraulic conductivity at larger values of the tension. It turns out that Dr. Rockhold had also proposed the same model for the anisotropy in the conductivity, providing a relatively clear basis for collaboration. The conceptual basis for the model follows.
Conceptual Basis for Subsurface Model

The sediments in the vicinity of the vadose zone transport field study (Gee and Ward, 2001; Ward et al., 2000) consist principally of sand with interstitial silt and silt beds (Last and Caldwell, 2001; Last et al., 2001). Thus a compound upscaling procedure, from the pore size to the sample size and from the sample to the formation size must be applied. In such a compound upscaling procedure it will be possible to assign some of the system to finer sediments, some to sand, and use known Hanford type soils and an assumption of constant matric potential $\psi = -h$ through the system to find the expected variability of moisture contents and thus the relative $K$ values (as functions of saturation, Hunt and Gee, 2002a) of sand and silts. These two components of the compound medium will be treated in a loose analogy to the particle and pore space of the Rieu and Sposito (1991) model, though of course here both portions have finite $K$. Using the relative occurrence of the ratio of these two $K$ values it will be possible to predict the anisotropy as a function of $\psi$ or of $\theta$.

The data consisted of some approximate maps of moisture contents in the subsurface as well as particle size data and water retention curves for 53 soils. Direct representation of $K$ as a function of saturation or tension for soils relevant to the project is missing, though information regarding $K$ under saturated conditions is available. The soil moisture maps allow determination of the length parameters associated with the Rieu and Sposito (1991) model, while the second portion of the data would, in principle, allow the determination of the relevant fractal dimensionality and equivalent porosity (fraction of high $K$ sediments), if there were an established protocol for assigning individual soils to the two soil types. The next step was intended to predict $K(S)$ and $K(h)$ from the treatment of Hunt and Gee (2002a) and compare with experimental data for these quantities. This protocol was intended to be developed based on the fraction of fine components in the soils: with the finer soils treated as silt loams (analogously to the McGee Ranch soil of Rockhold et al., 1988), and the coarser soils like the Injection Test Site (Sisson and Lu, 1984), both of which are typically monomodal. However, the PSD for many of the 53 soils are bimodal (or multimodal), and water retention data for these are not necessarily consistent with the hypothesis that soils with the finer components necessarily have the larger $K$ values at low moisture contents (high tensions) (the effects of the fines do not even show up). For bimodal particle size distributions the gap between the two modes of the distribution tends to produce a region of tensions with extremely rapid diminution of $K$ (Hunt and Gee 2002) and a lack of equilibrium drainage (Hunt and Skinner, 2005). The particular soil from the 1988 study that exhibited this behavior was called the North Caisson soil, and the rapid drop in $K$, as found in field data (Rockhold et al., 1988) set on already at a tension of about 25cm. On the other hand (Hunt and Skinner, 2005), the nonequilibriu conditions that result from such rapid drops in the equilibrium value of $K$ with increasing tension can (surprisingly) yield a higher measured value of $K$. Finally, it is true that almost 10% of the soils do have a unimodal particle size distribution analogous to the McGee Ranch soil, with a corresponding large value of an air-entry tension that could lead to these soils having a large $K$ at low moisture contents (higher tension values). Thus some of the soils that will tend to trap water at higher tension values will have a large $K$ value, but this fraction appears to be only about 20% of such soils, or about 10% of the
total. It is a valid question to ask whether that is a high enough percentage to develop sufficient connectivity (the fundamental topic of percolation theory) to allow the high $K$ regions to connect, even for limited distances in the horizontal direction.

The questions thus still to be answered are:

1) Can we use existing experimental evidence (summarized mainly in van-Genuchten parameters) to infer the $K$-values of the soils with higher fine components? This choice would be discouraged on account of the inconsistency of using a phenomenology to make predictions with a theory, at least if there is any alternative.

2) In the absence of direct experimental inference for $K$ is it possible to infer basic information regarding $K$ as a function of saturation from the water retention and particle-size data? This alternative seems better, although it carries the risk that there is only indirect verification of the validity for the prediction of the flow parameters. In particular, when the predicted hydraulic conductivity would reach a low enough value (typically on the order of 0.00000008 cm/s) the time scale for experimental equilibrium exceeds about six weeks (Hunt and Gee, 2002a), and the water retention data no longer reflect the pore size distribution, being limited by the flow rates of the water (Hunt and Skinner, 2005). The recognizable signal in the water retention curve is that it suddenly requires much higher tensions to remove significant amounts of water from the system.

3. What is then the fraction of water-retaining fine soils, which also have a larger $K$, and is this fraction consistent with percolation?

Because the results of the preliminary study cast some doubt on the initial interpretation, there is a need to collect additional data as well as to consult in detail with PNNL colleagues regarding the potential uncertainties in the modeling proposed.

**Present Theoretical Development**

In the proposal, the following results for the (unnormalized) expressions for $W(g)$ (the pdf for measuring a value of hydraulic conductance, $g$), were given

$$W(g) \propto \ln \left[ \left( \frac{L}{I + \epsilon^{1/2}} \right)^{1/2} \frac{g_c}{(1-D/3)(g-g_c)} \right] \quad g > g_c$$

and

$$W(g) \propto \ln \left[ \left( \frac{L}{I + x} \right)^{1/2} \frac{g_c}{(1-D/3)(g_c-g)} \right] \quad g < g_c$$

with $L$ a separation of water flow paths, $I$ the separation of critical, rate-limiting, values, $g_c$ of the hydraulic conductance, $g$, on these paths, $D$ the fractal dimensionality of the RS model, and $x$ the system size (linear dimension). The linearization that led to these equations was somewhat oversimplified; details are not incorporated here, but a result which is reliable over the full parameter space replaces $1/[(1-g/g_c)(1-D/3)]$ with $1/[1-$
This small theoretical change required that relevant results be obtained numerically. The numerical procedure was complicated by the logarithmic singularity in the distribution. Now comparison of the predicted distribution of $K$ values with experimental systems leads to Fig. 1 below, where the dashed lines indicate the values between which 68% of the experimental values are expected to lie.

![Log K vs. log V](image)

**Fig. 1** Log $K$, as function of experimental volume, $V$, compared with scale dependent $K$ from anisotropic fracture networks in a carbonate aquifer (Schulze-Makuch, 1996).

It is important to note that although the initial fit of the expected $K$ value as a function of system size was not unique, the same parameters were used to calculate the bounds on $K$. The experimental results from Fig. 1 were obtained from a fractured carbonate aquifer in Wisconsin (Schulze-Makuch, 1996). The value $\log(V)=0$ corresponds to the fundamental length scale in the problem. In both the fractured carbonate aquifer (Schulze-Makuch, 1996) and the Hanford subsurface, this value appears to be approximately 1 meter (pages 4.10-4.13 of Gee and Ward, 2001, aside from possible issues with experimental resolution). The ratio of the maximum to minimum $K$ in the Hanford subsurface is much smaller than that depicted; instead of about eight orders of magnitude, it will be closer to two orders of magnitude at larger tensions, whereas at smaller values of the tension (near saturation) it is likely to be 1 (a flat, or constant, graph) (internal Hanford studies as well as Zhang et al., 2003). This ratio is also very nearly the maximum anisotropy of the system. Thus, provided the original model is correct, we already have an excellent idea of
the spatial (and temporal) scales relevant to the Tc spill, i.e., when the spill has spread less than 1000m³ the effective $K_v$ value should increase to near the effective $K_h$ value.

**Results of Analysis of Vadose Zone Transport Field Study Soils**

In this section we present particle-size data of three types of soils from the Vadose Zone Transport Field Study site. One is a coarse sand similar to the soils from the Sisson and Lu study site (1984) later called (Hunt and Gee, 2002b) Injection Test Site soils (after Freeman, 1995). We will show that two of these soil types are very similar to soils from the Rockhold et al. 1988 study, which were called the McGee Ranch soil (a silt loam) and the North Caisson soil (a sand with a few percent silt, but for which the silt fraction occurred in a size range much smaller than the sand, i.e. a bimodal soil).

**Figure 1** Particle-size data for McGee Ranch silt loam (from Hunt and Gee, 2002a)

![Particle-size data for McGee Ranch silt loam](image)

**Fig. 2** The hydraulic conductivity of the McGee Ranch soil as a function of tension, h. Note that K remains relatively large out to large h values.
Fig. 3 Composite of North Caisson soil particle size distribution. Note the bimodality of the distribution as evidenced by the flat region in the middle.

Fig. 4 $K$ as a function of matric potential for North Caisson soil. Note that for tensions of 40cm, $K$ is already nearly an order of magnitude smaller than that for the McGee Ranch soil at tensions of 300cm.
Figs. 5-9 Some representative particle-size distributions from the Vadose Zone Transport Field Study site. Note that the first three soils have bimodal distributions like the North Caisson soil, but the last two are monomodal, like the McGee Ranch soil.
soil 57

soil 5

References


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