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Transport Processes Investigation: A Necessary First Step in Site Scale Characterization Plans

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ABSTRACT

We propose an approach, which we call the Transport Processes Investigation or TPI, to identify and verify site-scale transport processes and their controls. The TPI aids in the formulation of an accurate conceptual model of flow and transport, an essential first step in the development of a cost effective site characterization strategy. The TPI is demonstrated in the highly complex vadose zone in glacial tills underlying the Fernald Environmental Remediation Project (FEMP) in Fernald, Ohio. As a result of the TPI, we identify and verify the pertinent flow processes and their controls, such as extensive macropore and fracture flow through layered clays, which must be included in an accurate conceptual model of site-scale contaminant transport. We are able to conclude that the classical modeling and sampling methods commonly employed in most site characterization programs will be insufficient to characterize contaminant concentrations or distributions at contaminated or hazardous waste facilities sited in such media.

INTRODUCTION

Fundamental to any effective compliance or remediation strategy at a contaminated site is the formulation of an accurate predictive model of contaminant transport and groundwater flow. All successful predictive models must in turn be based upon a valid conceptual model. In practice, conceptual models are often based on general simplifying assumptions of standard saturated and unsaturated flow rather than site specific transport processes and their physical, chemical, geologic, and biologic controls. Predictive models based on these simplistic, generic conceptual models have consistently failed to correctly predict contaminant distribution, even when apparently conservative constraints and parameters have been incorporated (1). As a result, compliance and remediation strategies can fail with associated large remediation costs.

To avoid such failure, we propose that any site characterization or remediation strategy begin with an investigation of the transport processes and their controls active at a site. The TPI integrates information from a sequence of site specific activities in order to maximize the validity of conceptual models, yielding enhanced efficiency and cost effectiveness in compliance and remediation strategies. The TPI incorporates three main steps:

- **An exploratory phase** identifies probable hydrostratigraphic units and their degree of connectivity by integrating a search of the literature and existing site data with small scale field tests and exploratory surveys of local outcrops. Site specific transport processes and associated controls that must be investigated in order to yield a minimal scientific understanding of contaminant movement are hypothesized, and an uncontaminated analog site to test these hypotheses is located.

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- **Testing of hypothesized transport processes and controls** is carried out by conducting appropriately designed and sited transport processes characterization tests (TPCTs). As necessary, the TPCTs employ techniques such as pump tests, infiltration and tracer studies, dye pulses and excavation, and detailed soil/rock sampling in order to thoroughly investigative the field-scale transport processes active at the site.

- **Analysis of TPCT hypotheses testing** confirms or refutes the efficacy of the hypothesized transport processes and determines whether the active transport processes at the site are adequately understood or if additional investigation is required.

Information gained from the TPI must be integrated with information from elsewhere at the site to develop a valid site wide conceptual model of contaminant transport used to guide site characterization activities. This is accomplished by mapping of local geology with an emphasis on the relation of physical characteristics to confirmed transport processes and controls in order to provide a data set adequate for geostatistical and hydrologic transport simulations. These simulations, conditioned on the results of the TPI and other studies, will properly reflect the active site transport proeesses and their controls. Without this initial study of the physical processes controlling contaminant transport, characterization and remediation efforts may proceed in an expensive and undirected mode governed only by previously discovered contamination and compliance mandates.

The near surface vadose zone of the Fernald site was chosen for the TPI demonstration because of its location within a glacial till typical of deposits covering much of the North American continent (2). Glacial till vadose zones contain complex media and transport processes that provide a challenging conceptual model test. In addition, soluble species of uranium have been identified during surface based testing (3) and have been found at depth (4). Current conceptual models do not provide any reasonable mechanisms for the presence of uranium at depth, implying that unantieipated transport processes are active. In this paper, we present our demonstration of the TPI conducted at Fernald, Ohio as part of the DOE Technology Development Landfill Waste Area program.

EXPLORATORY PHASE

The exploratory phase integrated information gained from a literature search, several small preliminary tests (5), and a survey of local outcrops. The literature search identified transport processes and controls common in glacial tills. In this type of environment, macropore or fracture flow may define fast transport pathways (6, 7, 8, 9, 10, and 11). Media heterogeneity may also create zones of preferential flow (12, 13, and 14). Transport through the matrix of massive clays is likely controlled by diffusive mechanisms (2).

Paddys Run Creek, an intermittent stream crossing the Fernald site, afforded 2.5 km of stream cut bank for a survey of local outcrops. Stratigraphic units at these outcrops were investigated and mapped. Active seeps were noted in structural features (thrust faults) and at topographically low contacts at clay units with sand and gravel channels. Small scale dye infiltration tests were conducted in several outcrops that identified transport processes such as fracture and macropore flow. This exploratory phase suggested a connection among transport processes identified by the small scale dye transport tests, media heterogeniety, and larger scale geologic structural features.

Hydrostratigraphy and hypothesized flow processes and controls

Uranium is the most important contaminant at the site. Uranium transport may occur as soluble forms or as insoluble forms adsorbed to colloids. In either of these cases, aqueous subsurface transport processes are critical, and lithologic units were consequently defined on the basis of their hydrologic, rather than geologic, characteristics. Figure 1 presents six hydrostratigraphic units that were identified, each distinguished by unique hypothesized flow and transport processes and controls.

Unit 5 is the uppermost unit and contains the topsoil and silty loess comprising the bulk of the rooted zone. It was hypothesized that flow through this unit would be primarily vertical and controlled by plant material, root holes, and wormholes. Unit 4 consists of blocky structured silty clays and sandy silts apparently derived from paleosols. Flow in this unit was thought to be primarily vertical along fracture networks and through the matrix. Unit 3 is composed of bedded and fractured oxidized elays and silty clays. Both horizontal and vertical flow were expected to be possible in this unit, horizontally along bedding planes and laminae, and vertically downward through fractures. Unit 2 is composed of layered and bedded unoxidized clays. It was anticipated that horizontal and vertical fractures and bedding planes provide potential avenues for fast transport and preferential flow. Unit 1, the lowermost unit investigated, is composed of massive unoxidized clay with frequent and irregular gravels, pebbles, and cobbles. This "gray clay" has often been modeled as a barrier to vertical flow in the Fernald area.

Unit 6 is composed of sands and gravels. In general, the matrix is loosely compacted and was thought to provide the most transmissive avenue for contaminant transport of any unit. The relation of Unit 6 to the other units was varied and its sand stringers or channels were noted penetrating all other hydrostratigraphic units with the exception of the topsoils (Unit 5). Sand and gravel channels, as identified in outcrop, were hypothesized to provide possible avenues for transport through the massive gray clay (Unit 1).

Analog site selection

To efficiently investigate the anticipated flow processes at the contaminated site, an uncontaminated test site must be located that will provide an analog with respect to the active transport processes and controls that occur at the contaminated site. The analog site should fulfill the following criteria. It should be uncontaminated so that investigations may be conducted without causing contaminant migration or increased procedural burden. It should be nearby so that proximity provides an adequate analog (climate, weather, geology, etc.). Nearby outcrops for geologic mapping and interpretation are necessary, and ownership is desirable. Implementation of these selection criteria for our demonstration yielded the target selection of FEMP owned land along the west side of Paddys Run creek. This approximately 90 hectare uncontaminated field, used for dairy cow pasture, is within 500 meters of the main processing facilities and within 250 meters of several known locations of uranium contamination. All of these locations are on the East side of Paddys Run. Dissection of the local glacial till to depths of 6 meters by Paddys Run and the high relative topographic elevation of the field suggested hydrologic separation of the near surface from contaminated sites. Dissection also afforded approximately 2.5 km of stream cut outcrops ranging in elevation from 1 to 8 meters for geologic mapping.

Instrumentation
Infiltration rates, the pressure field, Cl^- tracer concentration fields, and rainfall were monitored by instrument and sampling arrays throughout the tests. The 10 meter wide, bi-level radial design of the array (Figures 1 and 2) acquires both three dimensional pressure and solute concentration fields, allowing a sensor derived interpretation of the flow field emanating from the infiltrometer and identification of transport pathways.

The size and design of the instrument arrays and sampling points were intended to allow spatially defined measurements and still ensure interception of field-scale transport pathways. Each instrument array contained forty-four sampling points with each sampling point containing a tensiometer, soil water samplers, and a Time Domain Reflectometry (TDR) probe in a sand filled instrument pack. The dimensions of the sampling point instrument packs (50 cm length by 10 cm diameter) were intended to afford interception of the anticipated macropore and fracture network without requiring integration of results over such large volumes as to render spatial definition of flow paths impossible. The use of sand in the instrument packs provided a high conductivity medium to facilitate interception of macropores. The elevation of the upper and lower sampling levels was determined by the stratigraphy encountered during sampling at each site. At the Clay Site, the upper levels were contained in the layered clays (Units 2 or 3) approximately 2 meters below ground surface, with the lower levels located at the upper surface of the massive unoxidized clay (Unit 1) at approximately 3 meters depth. At the Sand Site, the upper level instrument packs were located in the blocky structured silty clays (Unit 4) at approximately 1.1 meters depth, while the lower level packs were contained in thc underlying sands and gravels (Unit 6) at approximately 2.1 meters depth.

The pressure field was monitored by transducer equipped tensiometers at each of the forty-four sampling points at each site. Measurements were collected with a datalogger controlled, automatic acquisition system at fifteen minute intervals throughout the test. Transducers monitored by this system also supplied rain gage and infiltration rate measurements, also at fifteen minute intervals.

Tracer concentration fields were monitored by both passive and active means. Soil water samples for laboratory Cl^- tracer analysis were collected on one per day to three per day sampling schedules. TDR probes provided passive monitoring of solute concentrations at fifteen minute to one hour intervals over a multiplexed automated data acquisition system. Soil moisture was not of prime importance under the saturated conditions generated by the ponded infiltration, and TDR soil moisture calculations were made only to determine if saturated conditions did indeed exist in the sampling point instrument packs.

Infiltration and chloride tracer application
Infiltration was initiated at both the Clay and Sand Sites in mid June by pouring water into the circular infiltrometers and simultaneously opening the water supply valves, creating an essentially instantaneous ponded head of 5 cm. Infiltration was terminated at both sites in late August, 66 days after the start of the TPCTs.

Six days after the start of infiltration, an approximately 0.1M $CaCl_2$ tracer pulse was applied through the Clay Site infiltrometer and changing salinity levels were monitored by passive TDR probes and active soil water sampling at the forty-four locations at this TPCT site. On the following day, the same procedure was followed for the Sand Site. The Cl^- tracer pulse at the Clay Site was terminated after infiltration of 1,534 liters in 48 hours. The total Cl^- pulse at the Sand Site was

1,209 liters in 53 hours. The lower volume infiltrated at the Sand Site was due to a lower infiltration rate through the Sand Site infiltrometer surface, which at the time was less conductive than at the Clay Site.

Dye Pulse and Excavation of the Clay Site

After 61 days of infiltration, FD&C Red #3 dye was added to the Clay Site infiltrometer supply tanks. Four days later, after infiltration of 400 gallons of dye, the dye tracer pulse was terminated. At the same time, clean water infiltration was halted at the Sand Site.

Excavation of the Clay Site down to and including the lowermost massive gray clay (Unit 1) was accomplished using both a large back hoe and hand tools. Use of the back hoe allowed rapid removal of one foot to two foot layers. The subsequent hand tool examination ensured that complete and detailed transport features would be revealed.

ANALYSIS OF TPCT HYPOTHESES TESTING

Results from each type of measurement obtained during the TPCTs confirmed the validity of our hypothesized transport processes and yielded a much improved understanding of their active controls at the Fernald site. We are not able to present all results in detail in this paper.

Infiltration

The long time, steady-state infiltration rate should approximate the saturated hydraulic conductivity of the least conductive media being infiltrated (15). At the Sand Site (Figure 1), the limiting media would be silts and loess with saturated conductivities ranging from 10^{-5} cm/s to 10^{-7} cm/s (16). At the Clay Site, the limiting media would be the massive clay (Figure 1) with conductivities ranging from 10^{-7} cm/s to 10^{-11} cm/s (16). Instead, infiltration rates at both sites approached 10^{-3} cm/s, exceeding the limiting conductivities by two to eight orders of magnitude and suggesting that infiltration was controlled by the capacity of the conducting macropore system through and below the infiltrometer surface.

Over the sixty-six day infiltration period, 61,000 liters of water were infiltrated through the system at the Clay Site and 78,000 liters of water at the Sand Site. Neither site became saturated at the surface anywhere outside the infiltrometer, also implying an interconnected subsurface transport-pathway system capable of significant transport.

Pressure head field

The pressure head field across the 3-D instrument array showed a general pattern of mounding beneath the infiltrometer, with local head perturbations of -10 to -5 cm distinguishing areas of preferential flow. Most sampling points showed rapid response to rainfall events (e.g., approximately 30 minutes at 3 meters depth), indicating that the majority of near subsurface media was well connected to the surface by a macropore system. The rise in head was typically almost ten times the amount of precipitation. However, some sampling points displayed neither rapid nor large rises in head, suggesting that the macropore and fracture system did not connect the entire investigated volume.

Tracer concentration field

Based on soil water samples and TDR measurement, overall tracer movement at both sites was initially vertically downward below the infiltrometer, then horizontally. Horizontal distribution of tracer concentrations began sooner at the Clay Site than at the Sand Site and dissipated almost ten days sooner. The distribution of tracer concentrations within the generally vertical and radial flow field was highly heterogeneous at the Clay Site and flow appeared to follow definite pathways (Figure 2). Arrival times of peak concentrations took less than one hour at some sampling points in the Clay Site and affirmed our hypothesis of rapid transport pathways. Arrival times of peak concentrations exceeded those calculated solely on the basis of bulk media properties by four to eight orders of magnitude. Contributions of multiple transport pathways at some sampling points were observed as multiple peaks in breakthrough curves (Figure 3).

Dye Pulse and Excavation

The dye pulse and subsequent excavation of the Clay Site presented striking visualization of flow processes and transport pathways, and confirmed data from the instrument array (Figure 4). Dye capture was prevalent in the uppermost layer of topsoils and loess (hydrostratigraphic Unit 5), while vertical root, worm, and insect macropores and fractures transported the dye tracer through the underlying blocky structured clays (Unit 4). The matrix of this unit was stained only in the immediate vicinity of macropore flow and transmissive fractures. Macropores were noticeably missing from sand and gravel media, likely because the unconsolidated matrix does not support pore walls just as it limited instrument borehole depth (see **TPCT site selection** above). As hypothesized, horizontal dye movement along horizontal fractures and bedding planes was noted both in the oxidized bedded clays (Unit 3) and in the unoxidized bedded clays (Unit 2).

In addition to these expected features we found tubular pathways approximately 2 cm by 3 cm in cross sectional area meandering from just beneath the topsoil down to depths well below the upper surface of perched water tables as defined by standing water in boreholes during installation in late May. These large macropores were later identified as crayfish burrows that are endemic to the area. Crayfish burrows and burrow complexes filled with dye connected the topsoils to fracture networks in the bedded clays. These pathways are each capable of transmitting more than the maximum flux observed passing through the infiltrometers. The large incidence of dye stained crayfish complexes in the southern portion of the site (Figure 4), none intercepted by sampling points, may explain the lack of tracer concentrations sensed in that area by the instrument array. At the Clay Site, these and all other bioturbated fast transport pathways terminated in the unoxidized layered clays (Unit 2).

Sand and gravel stringers and channels were observed at all depths and penetrated even the massive gray clays (Unit 1). Dye transport through numerous silty sand zones was discovered in the layered clays. Unfortunately, at the depths where Unit 1 was encountered (3 to 5 meters), the dye pulse was diluted to an extent that no confident association could be made between the infiltrated dye pulse and flow through sand stringers within the massive gray clay.

CONCLUSIONS

Transport processes, controls and connectivity

The Transport Process Investigation was effective in identifying pertinent flow processes and controls. We can conclude that a conceptual model for the near surface vadose zone at the Fernald site must incorporate the following processes and controls. The dominant flow and transport pathways through the topsoil to the bedded clays are vertical bioturbated macropores and fractures. These pathways are capable of rapid and abundant transport during most of the year, when essentially saturated conditions exist or when rainfall events are of sufficient magnitude to enter the macropore system. The bedded clays support a pervasive network of transmissive horizontal and vertical fractures and are connected to the surface by vertical fractures and macropores. Some crayfish burrows are continuous from the surface to the bedded clays and intersect the fracture network in the clays. Crayfish burrows may provide the dominant transport pathways in some areas, allowing contaminants to bypass extensive volumes of the near surface media.

Channels and stringers of sands and gravels provide localized zones of preferential flow and are well interconnected by fractures and macropores. In this extensively reworked and deformed glacial depositional environment, highly transmissive sand and gravel channels and shear zone fractures penetrate portions of the otherwise impervious massive gray clays, calling into doubt its effectiveness as a barrier to flow and transport. The seeps and preferential flow features noted in the exploratory geologic survey may be representative of field-scale transport pathways. Trends in infiltration rates and changes in the pressure field indicate that the system is well drained, with a significant potential for contaminant transport.

Further studies

The connectivity of transport pathways from the surface to depth has been implied at the field scale by the well drained nature of the flow system and should be fully investigated by larger scale tracer or pump tests. Larger scale tests will require that the hydrologic separation of contaminated and uncontaminated sites be conclusively demonstrated. Tests also must be conducted to conclusively determine if the deposits of massive gray clay are effectively breached by discontinuities and penetrations by highly transmissive sand and gravel channels.

Implications for site characterization strategies

Through implementation of the Transport Processes Investigation we were able to distinguish the governing transport processes and controls active in this highly complex near surface glacial till and identify those that must be included in a defensible conceptual model for contaminant transport at the site. Site characterization plans formulated by classical statistics and sampling strategies, and predicated on generic conceptual models of subsurface flow and transport, will likely not recognize the potentially rapid transport processes and controls identified by the TPI. Consequently, such generic conceptual models will characterize contaminant concentrations and distributions only by chance or through prohibitively extensive and expensive classical sampling plans.

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TESTING OF HYPOTHESIZED FLOW PROCESSES AND CONTROLS

Design of the Subsurfaee Transport Proeesses Charaeterization Test (TPCT)

Based on the information gained from the exploratory phase, we anticipated the dominant active transport processes would be macropore flow through insect, worm, and root holes; flow along bedding planes and through fractures; and preferential flow due to media heterogeneity. The most important heterogeneity was anticipated to be highly transmissive sand and gravel stringers and channels penetrating more restrictive layers.

To confirm or refute our hypotheses we designed TPCTs that would investigate the presumed transport processes and controls. Because saturated conditions were encountered during an early spring site survey and these conditions can exist for a three or four month period, and in order to provide the worst case conditions for colloidal or dissolved contaminant transport, the test was designed to simulate saturated conditions. A 2.2 meter diameter circular ponded surface infiltrometer was used to produce a steady-state saturated flow field for a fifty to one hundred day period. The pressure field was monitored in time and space throughout the test to allow definition of the overall flow field and to detect any preferential flow. To aid in identifying transport pathways, Cl^- tracer pulses were released into the flow system through the infiltrometers after saturated, steady-state conditions had been obtained. Near the end of the test and while still under steady-state saturated conditions, a pulse of FD&C Red #3 dye was introduced through the infiltrometer at one TPCT. The site was then excavated to identify dyed transport pathways and zones of preferential flow. The excavation also allowed inspection of hydrostratigraphic units and their local connections unaffected by weathering or erosional dynamics such as those present along Paddys Run. Site excavation allows us to confirm or refute interpretations derived from sensor and sampling analysis of the Cl^- tracer pulses and to visually relate transport pathways to the site geologic conditions and flow processes.

TPCT Site selection

After appropriate TPCTs have been designed, specific sites suitable for implementation must be located. Within the analog area west of Paddys Run, several candidate sites for the TPCTs were chosen during a preliminary survey in the early spring. An archeological survey was conducted at these locations and in late May a number of exploratory holes were augured. Based on samples from the exploratory holes and the hydrostratigraphic units and transport controls identified in the exploratory phase, two sites were chosen for TPCTs. The two sites contained distinct geologic sequences that together represented the major hydrostratigraphic units and transport process controls found in outcrop during the exploratory phase, allowing investigation of all hypothesized transport processes and their controls.

The Sand Site (Figure 1) was composed of topsoils and loess (Unit 5) overlying blocky structured clays (Unit 4). Beneath these units the only geologic unit encountered in any significant amount was a large, unconsolidated body of sand and gravels (Unit 6). This underlying sand and gravel unit could be penetrated only to a depth of approximately 2.5 meters before the borehole walls collapsed. Bedded clays (Units 2 and 3) were encountered at the Sand Site only in isolated samples above the sand and gravel unit.

The Clay Site (Figure 1) contains a hydrostratigraphic sequence including all of the units at the Sand Site plus horizontal beds of oxidized and unoxidzed clays (Units 3 and 2) extending across the site and overlying the massive unoxidized gray clays (Unit 1). The sands and gravels of Unit 6 were evidenced only in stringers and one sand channel, not in a large sand body as at the Sand Site.

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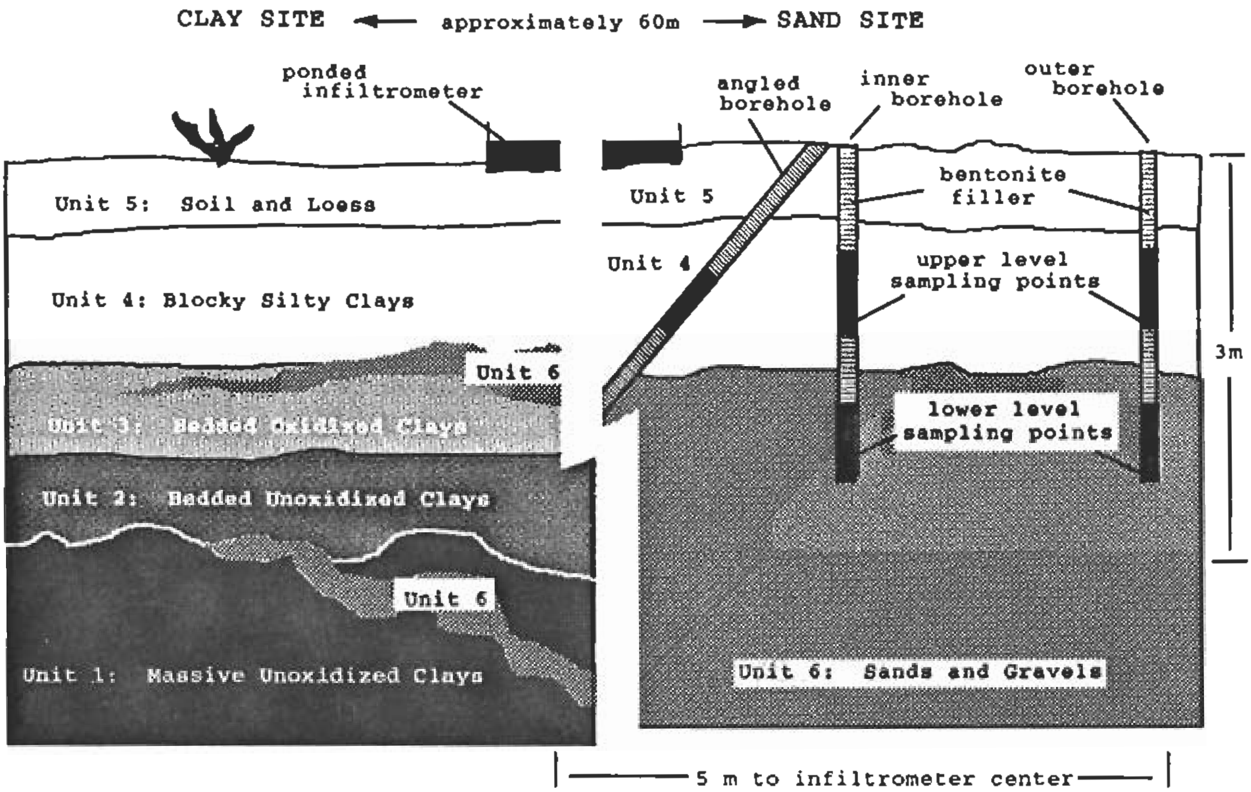

Figure 1. Hydrostratigraphic units at the "Clay" and "Sand" Sites. A cross section of the instrument array at the Sand Site is also shown. The instrument array at the Clay Site is identical except the upper and lower level instrument packs are approximately 1 m deeper, with the lower packs just above Unit 1.

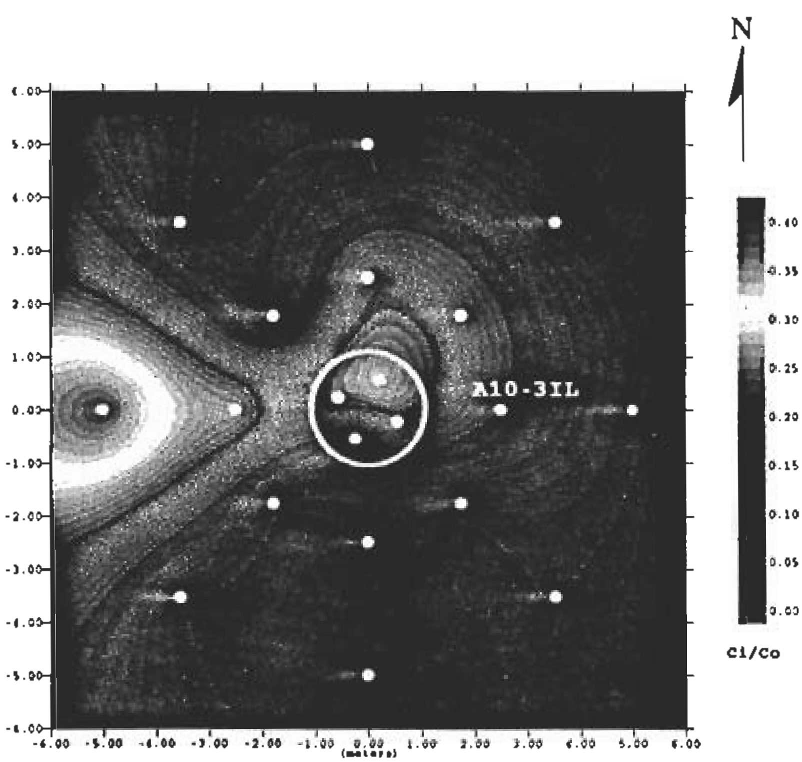

Figure 2. Cl^- tracer concentrations distribution at the lower (3 m) level of the Clay Site 119 hours after start of infiltration. White dots are operational sampling points. The 2.2 m diameter infiltrometer is centered in the figure.

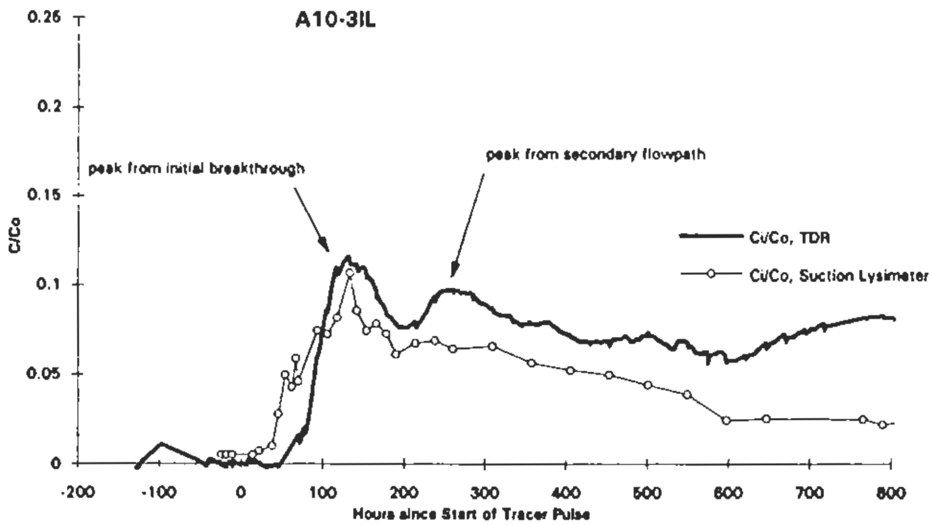

Figure 3. Repeated peaks in Cl^- tracer breakthrough curves due to contributions from multiple flowpaths. The sampling point is located 2.5 m East of the infiltrometer at a depth of 3 m (see Figure 2).

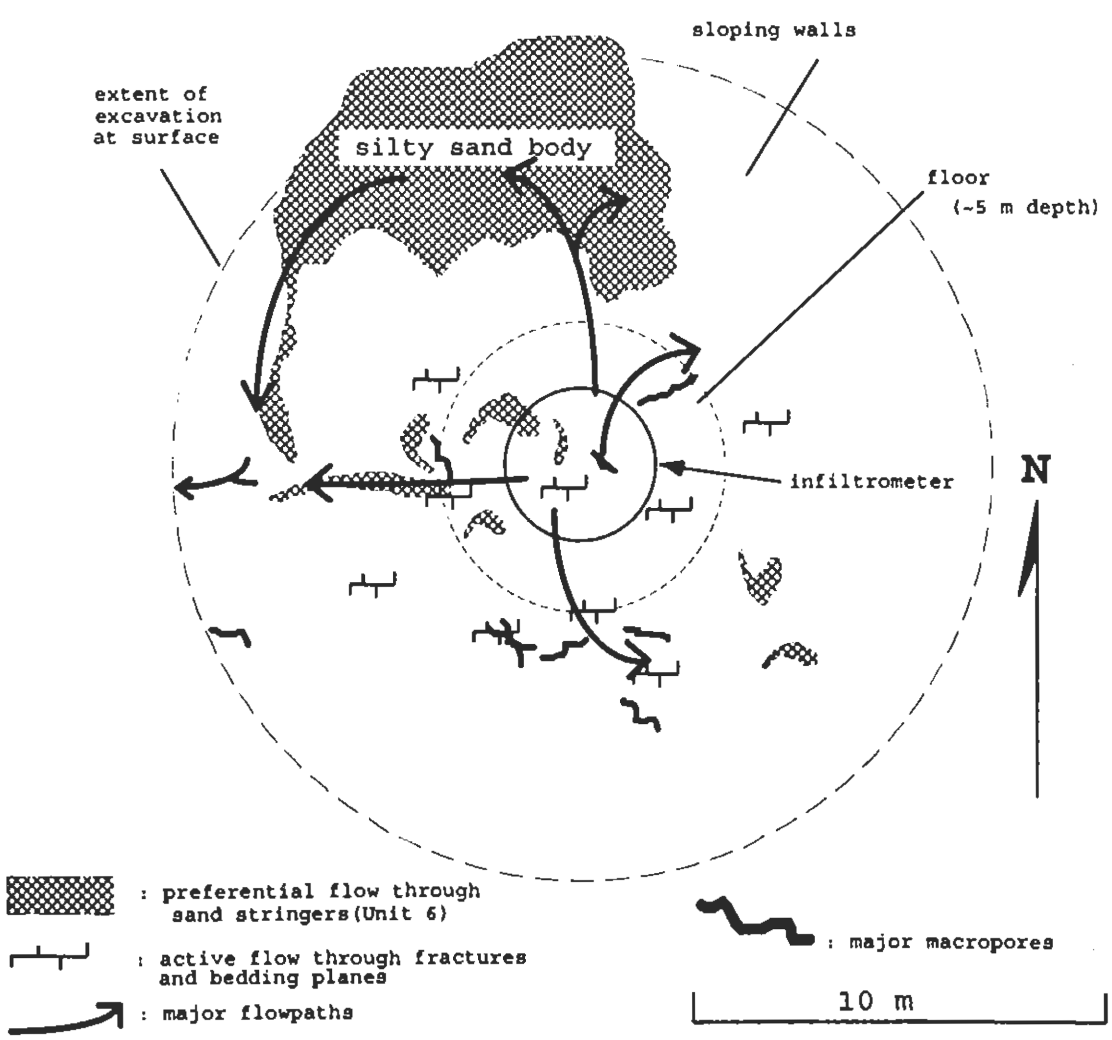

Figure 4. Plan view of major macropores, fractures, and zones of preferential flow found during excavation at the Clay Site. Noted features are integrated over the depth of the excavation.