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SEAMIST SOIL SAMPLING FOR TRITIATED WATER: FIRST YEAR'S RESULTS

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ABSTRACT

SEAMIST is a recently developed sampling system that enables one to measure various soil parameters by means of an inverted, removable, impermeable membrane tube inserted in a borehole. This membrane tube can have various measuring devices installed on it, such as gas ports, adsorbent pads, and electrical sensors. These membrane tubes are made of a laminated polymer. The Lawrence Livermore National Laboratory in Livermore, California, has installed two of these systems to monitor tritium in soil resulting from a leak in an underground storage tank. One tube is equipped with gas ports to sample soil vapor and the other with adsorbent pads to sample soil moisture. Borehole stability was maintained using either sand-filled or air-inflated tubes. Both system implementations yielded concentrations or activities that compared well with the measured concentrations of tritium in the soil taken during borehole construction. In addition, an analysis of the data suggests that both systems prevented the vertical migration of tritium in the boreholes. Also, a neutron probe was successfully used in a blank membrane inserted in one of the boreholes to monitor the moisture in the soil without exposing the probe to the tritium. The neutron log showed excellent agreement with the soil moisture content measured in soil samples taken during borehole construction. This paper describes the two SEAMIST systems used and presents sampling results and comparisons.

INTRODUCTION

A rotating target neutron source (RTNS) was installed at the Lawrence Livermore National Laboratory (LLNL) in Building 292 in 1977 and used until 1987. The purpose of the facility was to study the effect of high-energy neutrons on various materials that may be employed in fusion reactors. The target was composed of titanium tritide, which released a high-energy neutron when struck by a high energy deuteron. The effort was international in scale, attracting scientists interested in designing a source of "clean" electric power.

Tank 292-R1U1 was installed in 1974 to receive water used to rinse the floors in the experimental rooms. A leak in the tank was discovered in 1989. Subsequent soil samples taken near the tank showed tritium concentrations ranging from 10^8 picocuries per liter of soil moisture (pCi/L_{sm}) near the leak to 10^4 pCi/L_{sm} above the water table and approximately 40 ft below the leak. The concentrations were lower 8 ft northwest, reaching 10^7 pCi/L_{sm} maximum at 11 ft, and decreasing toward the water table to approximately 10^4 pCi/L_{sm} (Fig. 1). Other boreholes in the vicinity of the tank indicated far less or no tritium concentration.

The tritium in the first water bearing zone (approximately 50 ft thick) is being sampled and monitored with piezometers. The SEAMIST technology, which although unproven, is simple in concept and was selected to provide vadose zone monitoring capability over an extended period without constructing a new set of boreholes for each round of samples.

METHODS

SEAMIST is a borehole instrumentation system (patent pending) developed by Science and Engineering Associates (SEA), Inc. (Keller and Lowry, 1990). The system consists of a membrane tube that carries a sampling system into a borehole and a reel for removing that tube (Fig. 2). The tube is inserted into the borehole by means of an air pressurization system. It is retracted by a tether attached at one end to the bottom of the tube and attached to the reel at the other end. The membrane is held against the sides of the borehole by either air pressure, sand, or grout to prevent borehole collapse.

Two of the SEAMIST systems were deployed in 40-ft-deep boreholes near the leak site. The boreholes were drilled with a 4-in. bit making an effective diameter of 4.3 in. The SEAMIST systems installation were described by Lowry and Cremer (1991). One system was equipped with gas sampling ports installed in borehole B-16 and the other with adsorbent pads installed in borehole B-17.

The gas port system was installed near the leak site in borehole B-16. Eight ports were located at specified depths to collect soil vapors from different geological strata. The depths of these strata were identified from the geologic log of nearby borehole U-292-001 (Fig. 3). The sample membrane was constructed of a laminated vinyl/polyester material. Each port was composed of a screened disk on the outer surface of the membrane. The gas ports were connected to a base pipe at the surface by means of 1/16-in.-inside-diam (Tygothane) tubing. After emplacement, borehole stability was maintained by filling the membrane with sand, thus making the system semipermanent.

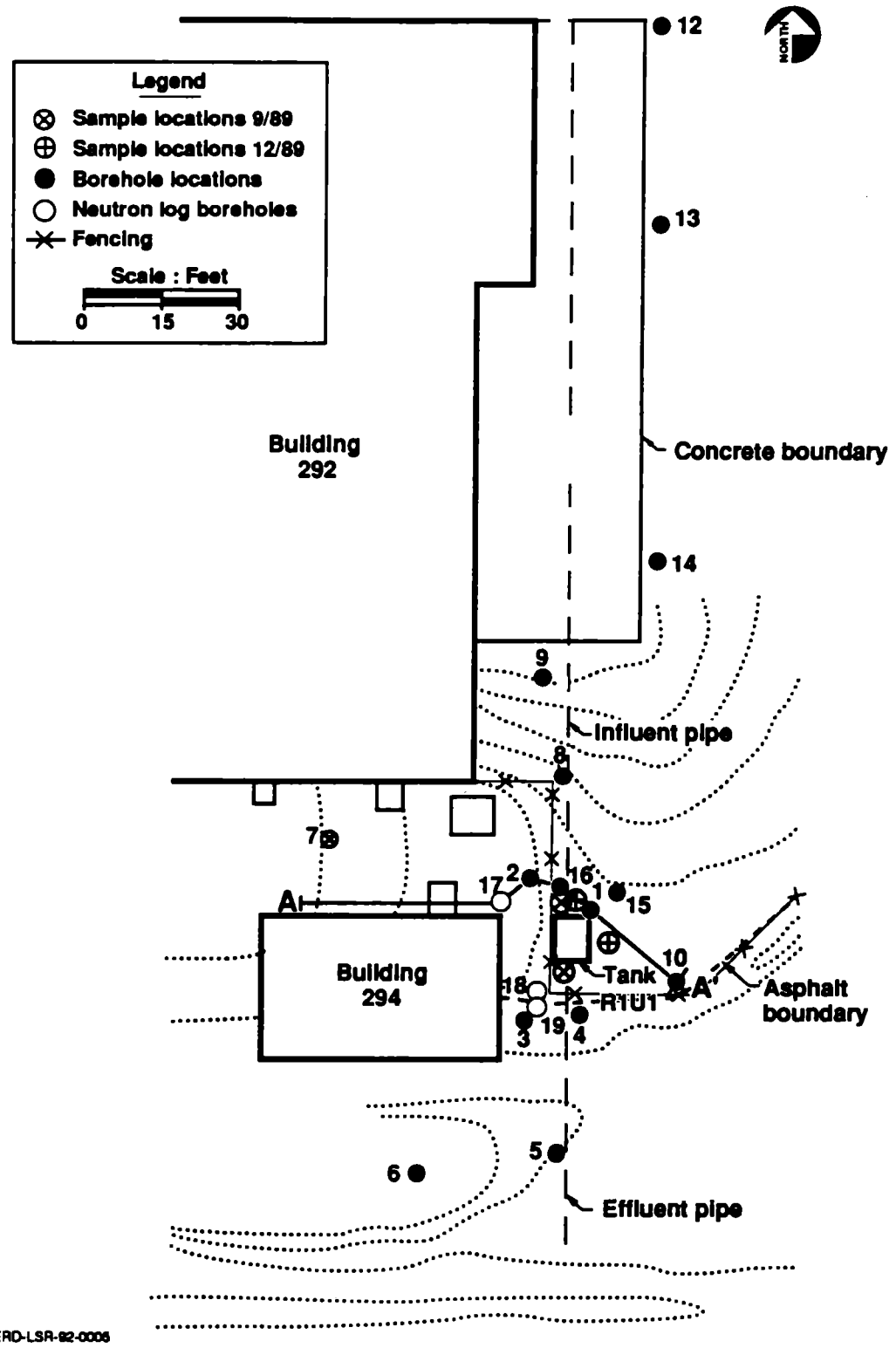

Figure 1. Borehole sample locations in Tank U292-R1U1 vicinity.

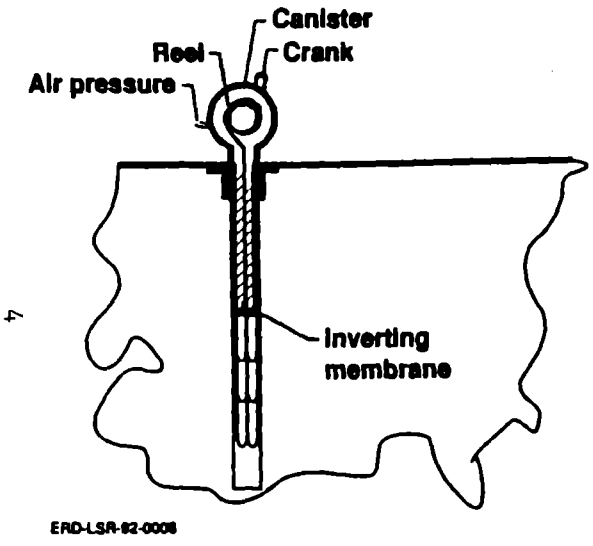

Figure 2. SEAMIST emplacement system.

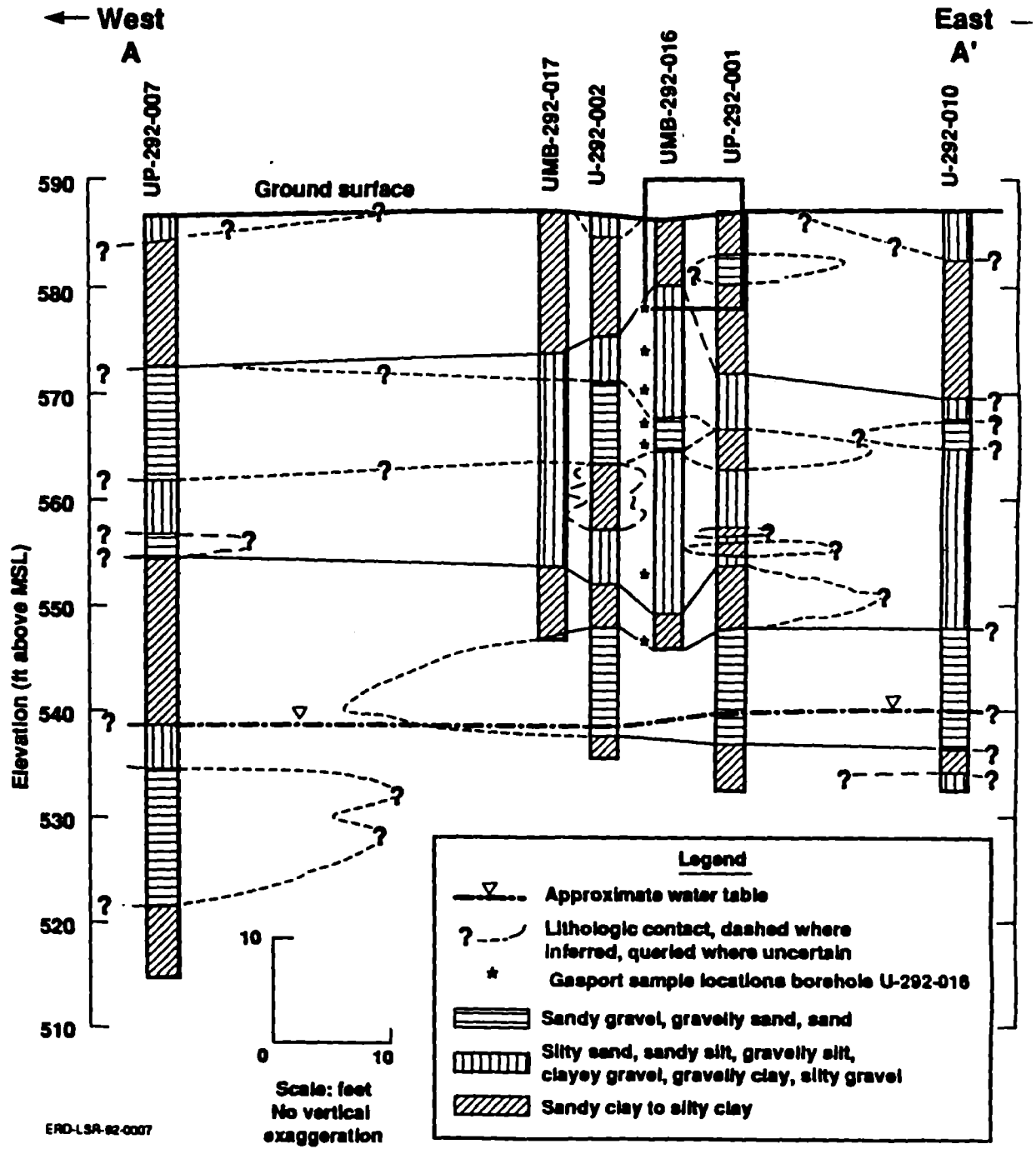

Figure 3. Cross section A-A' lithology of B-292 Area.

Soil gas samples were collected by applying a 23.6-kilopascal (KPa) vacuum to all ports simultaneously. Water vapor in the soil gas was condensed and frozen in the cold traps cooled with dry ice and isopropanol (Martins, 1992). A total flow rate of 8 L/min was maintained for the eight ports. The flow rate for the initial sampling varied from 0.9 L/min for the port closest to the surface to 0.6 L/min for the deepest port. The volumes of water collected after pumping for 4.5 h under these conditions are shown in Table 1.

Table 1. Liquid water volumes collected from the SEAMIST gas port system.

Depth (ft)	Amount (m/L)	Lithology
8.5	1.8	Sandy silt
13.5	1.5	Sandy silt
16.0	1.6	Clayey gravel
19.0	1.6	Sand
22.0	1.6	Sand
26.5	1.6	Silty sand
34.5	1.5	Silty sand
40.0	1.4	Sandy clay

The volumes of water collected appear to be dependant on depth rather than lithology. The September and November samples were collected under similar conditions. The water was analyzed for tritium content by scintillation counting.

The second system consisted of two parts: the sampling membrane and a blank to hold the borehole open between sampling events. Borehole stability was maintained by means of 0.3 to 1.0 psi air pressure in a blank membrane made of vinyl-laminated nylon material. When sampling was desired, this membrane was withdrawn onto its reel and an adsorbant collector sampling membrane of vinyl-laminated polyester material was substituted. Again, borehole stability was maintained with 0.3 to 1.0 psi air pressure. The adsorbant collector membrane had 20 pockets at 2-ft intervals built onto its exterior. Two polypropylene pads weighing a total of 5.0 g were placed in each of the pockets.

The collection system was removed 24 h after emplacement, and during that time the pads had collected approximately 0.4 g of water and some dirt. The pads were put in vials, the combination was weighed, and 20 mL of water was added to each vial. After 12 h, a sample of the water was taken and analyzed for tritium content by liquid scintillation counting. The pads and

vials were dried and reweighed, and the original weight of water in the pads was determined. The tritium content of this water was calculated.

The location of the absorber emplacement borehole was selected based on the sampling results from boreholes B-1 to B-10 shown in Figure 1. The highest concentrations were found in borehole B-1 near the leak site, and the next highest were found in borehole B-2, thus suggesting an east to west concentration gradient in the soil moisture. The location for B-17 was also selected because horizontal migration of tritiated water was suspected in this direction. The concentration profile after borehole construction is shown in the same format in Figure 4 as the lithology in Figure 3. The data are consistant with an east to west spread of tritiated water in the vadose zone.

The "absorber" SEAMIST borehole is used to monitor soil moisture movement. The soil moisture is recorded with a neutron logging tool (Campbell Pacific Nuclear Corporation, 503 Hydroprobe). Measurements of moisture content of soil samples taken during borehole construction were made by measuring the volume of the brass liner and determining the weight of the water by weighing the specimen and then drying it to constant weight at 105°C. The neutron log counts were calibrated against the volume percent moisture measurements obtained during the analysis for tritium in the soil moisture during borehole construction.

Some additional experiments with the gas port system were performed to determine the influence of suction applied to one port on those ports above and below it. It was suspected that surface air would be pulled down the borehole during collection. Therefore, one port was excluded and connected and a vacuum of 23.6 KPa was drawn on the other ports. The pressure drop was measured at the excluded port. In addition, air samples from each port were drawn and submitted for mass spectrometer analysis.

The tritium concentrations are expressed in picocuries per liter of soil moisture. The drinking water standard is 20,000 pCi/L. Tritium units (TU) are also sometimes used to express tritium concentrations in water. The conversion factor is 1 TU = 3.2 pCi/L.

RESULTS

Tables 2 and 3 show tritium concentrations measured in liquid extracted from soil samples taken during borehole construction and from samples collected from the two SEAMIST systems. These data are graphically represented in Figures 5 and 6. Examination of the two graphs shows excellent correlation between the May and June 1991 SEAMIST sample concentrations compared with the soil concentrations in May 1991.

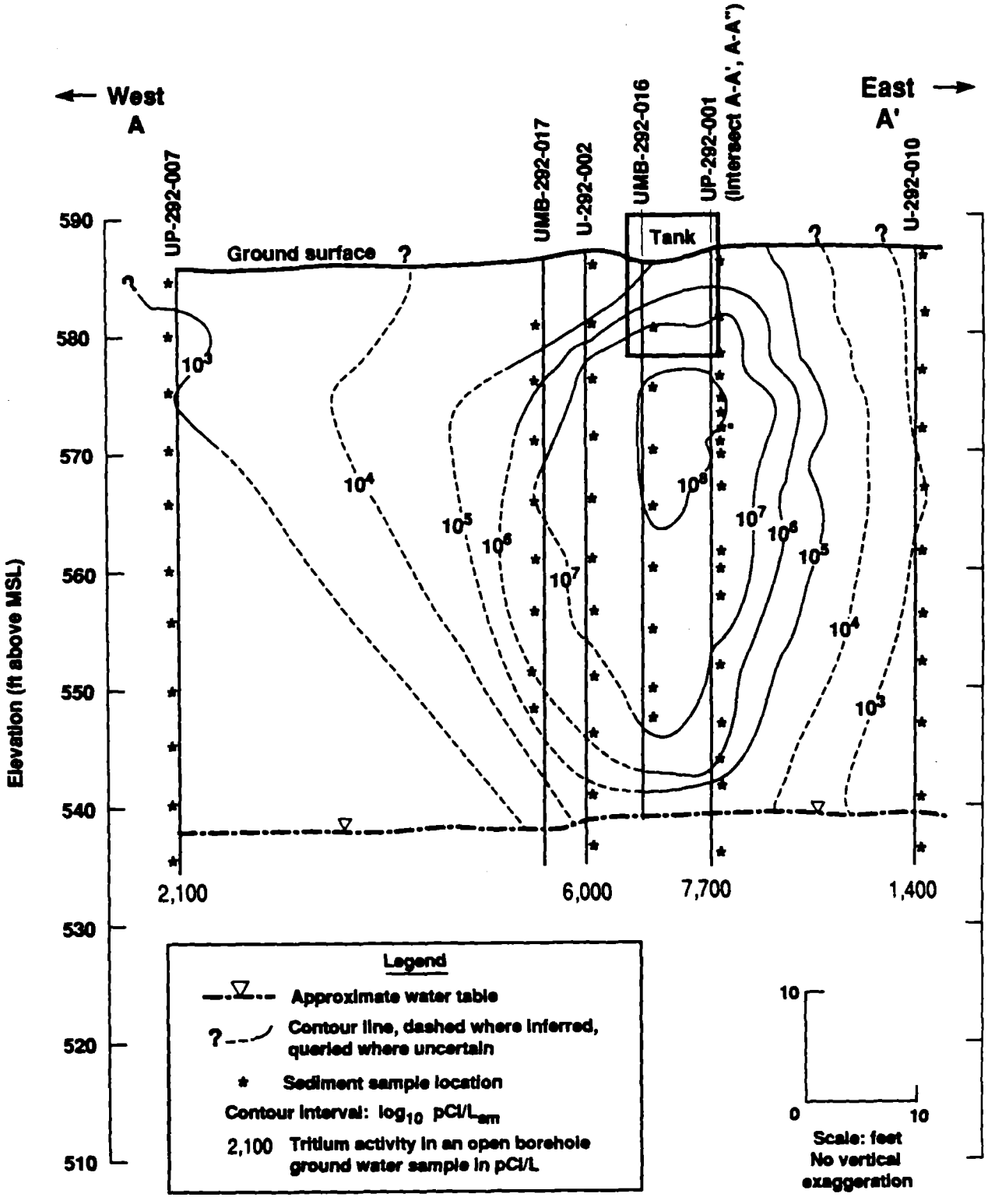

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Figure 4. Tritium concentrations on east-west fence diagram A-A'.

Table 2. Comparison of tritium in moisture concentrations of soil samples taken during borehole B-16 construction (drilled 5-16-91) with vapor concentrations taken with SEAMIST gas ports on 5-23, 8-08, and 11-18-91.

Depth (ft)	Lithology	May	August	November	Sample type
		---------- (10^6 pCi/L_{sm})[a]		----------	
6.0	Sandy silt	23			Soil
8.5	Sandy silt	62	56	45	Vapor
11.0	Sandy silt	135			Soil
13.5	Sandy silt	127	133	95	Vapor
16.0	Clayey gravel	143			Soil
16.0	Clayey gravel	123	129	79	Vapor
19.0	Sand	124	126	86	Vapor
21.0	Sand	123			Soil
22.0	Sand	78	34	48	Vapor
26.0	Sandy silt	37			Soil
26.5	Sandy silt	32	39	34	Vapor
31.0	Silty sand	24			Soil
34.5	Silty sand	22	31	20	Vapor
36.3	Silty sand	34			Soil
39.5	Sandy clay	16			Soil
40.0	Sandy clay	6.0	19	17	Vapor

[a] Picocuries per liter of soil moisture.

Table 3. Comparison of tritium in moisture concentrations from soil samples taken during borehole construction (drilled 5/17/91) with soil moisture concentrations extracted by SEAMIST adsorbent pads on 6-23, 9-20, and 12-06-91.

Depth (ft)	Lithology	May soil	June pads	September pads	December pads
		(10^6 pCi/L_{sm})[a]			
2.0			0.23	0.01	(b)
4.0			0.24	0.13	(b)
6.0	Sandy silt		0.25	0.16	(b)
6.5	Sandy silt	0.05			
8.0				0.3	0.4
10.0	Sandy silt	0.26	0.66	0.6	0.9
11.0	Sandy silt	1.3			
12.0	Sandy silt		1.9	1.6	2.3
14.0			3.1	3.3	4.3
16.0	Clayey gravel		5.2	6.6	6.2
16.5	Clayey gravel	6.2			9.1
18.0				8.3	8.3
20.0	Sand		12	14	16
21.5	Sand	12			
22.0	Sand		13	15	17
24.0			12	12	13
26.0	Silty sand	9.9	10	13	13
28.0				15	15
30.0	Silty sand		9.9	14	13
31.5	Silty sand	9.2			
32.0	Silty sand		8.2	10	12
34.0	Sandy clay	4.6	7.4	11	9.3
36.0	Sandy clay		4.5	4.9	5.4
36.5	Sandy clay	3.5			
38.0	Sandy clay			2.8	2.6
39.5	Sandy clay	0.86			
40.0	Sandy clay	0.71	0.65		0.9

[a]Picocurries per liter of soil moisture.

[b] Below limit of detection (0.001 x 10^6 pCi/L).

Figure 5 shows the results of the soil vapor collection system compared to the soil moisture concentrations in soil samples taken during borehole

construction. As mentioned above, the initial concentrations from the SEAMIST gas ports compare well with those of the moisture extracted from borehole soil samples. However, the November samples suggest a possible decrease in concentration from the May and August samples. Collection and analyses of February 1992 samples may help resolve the apparent decrease in concentration. Some rain water may have seeped into the borehole/membrane interface. This area was re-sealed to eliminate this possibility.

There also may be some transport of air down the borehole/membrane interface during sample collection. To measure the potential air flow down this interface, all ports but one were connected and a vacuum of 23.6 KPa was applied. The unconnected port showed a loss of pressure of less than 0.1 KPa. Each port was isolated in turn and the same result was achieved (Fig. 7). The results suggest that there is no significant communication between the ports.

Likewise the measurements of oxygen and carbon dioxide showed that air from the surface via borehole interface was not sampled. The oxygen content of the air at the earth's surface is expected to be 21%, whereas the amount found in the mass spectral analysis of the soil vapor was 17.5% or less. In addition, the oxygen and carbon dioxide content of the different depths sampled varied inversely with each other as shown in Figure 8.

Figure 6 shows the concentrations in the soil during borehole construction as compared to the concentrations collected with the pads 1, 4, and 7 months later. The initial results are similar for the soil and June adsorbant pads samples except near the surface. The December samples show no measurable tritium concentrations until the 8-ft depth, similar to the soil samples. Air leaking from the blank tube during the previous month may be causing the tritiated water to migrate upward. The first blank membrane was replaced with a new membrane in October. This membrane had a few much smaller air leaks. Patches were applied to make that membrane leakproof. SEA is now bubble testing membranes before use and is also working on a new passive membrane pressurizing system that will not require a continuous flow of air.

Although emplacement/recovery for sampling with the pads is somewhat more cumbersome than the vapor extraction with the gas port system, the pads system apparently yields an excellent direct monitoring of the soil moisture. Successive pads samples in September and December suggest an increase in tritium content at the 20- to 30-ft depth interval. Soil moisture content measurements are driest at the 20-ft depth, thus indicating vapor phase transport.

In addition, the open borehole pad system retains a flexibility which is lost in the semipermanent gas port installation as a result of filling the membrane with sand. Besides having the capability to change the sensors attached on the exterior of the

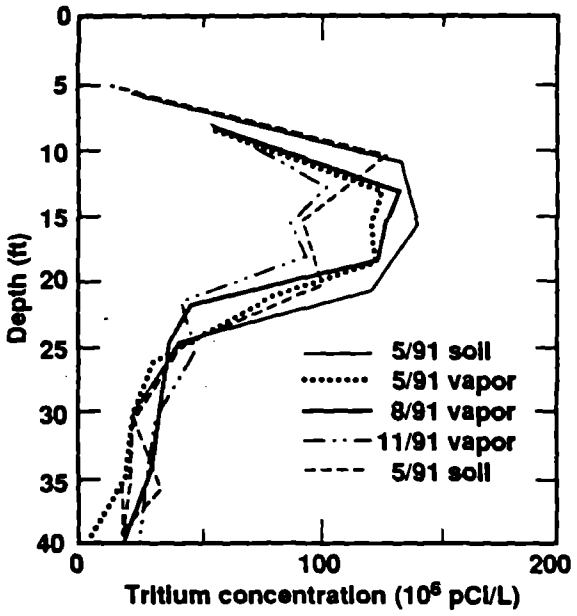

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Figure 5. Tritium concentration in soil moisture and SEAMIST vapor samples.

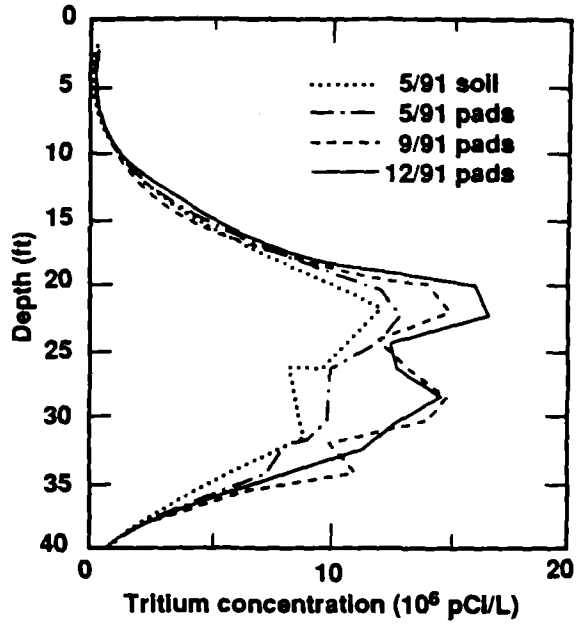

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Figure 6. Tritium concentration in soil moisture and SEAMIST pads moisture.

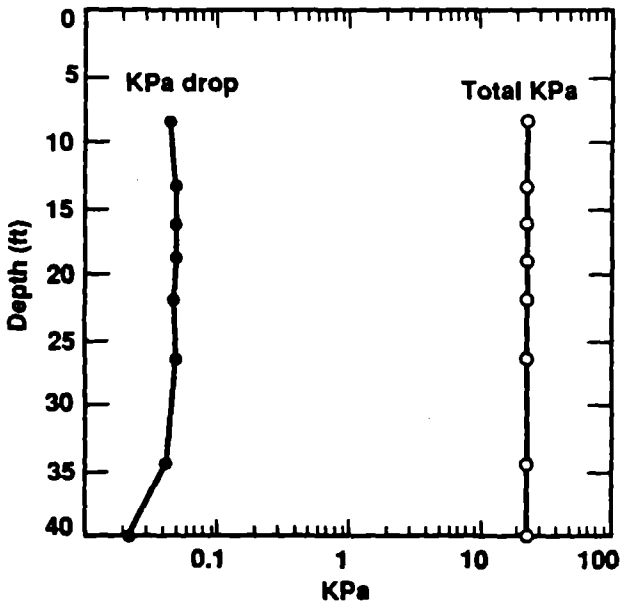

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Figure 7. Pressure drop at port when other ports are connected to 23.6 KPa vacuum.

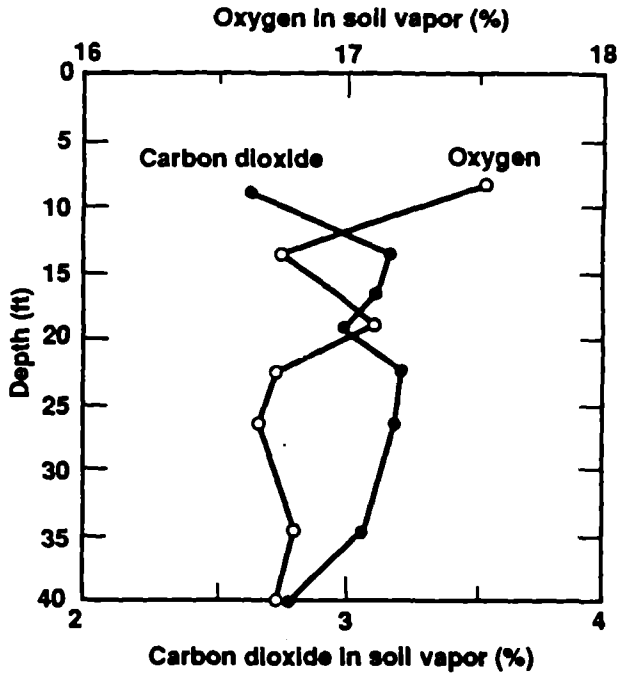

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Figure 8. Changes in oxygen and carbon dioxide concentrations with depth in borehole UMB-292-016.

membrane, an air-filled borehole blank was used to monitor moisture with a neutron logging tool. Initial neutron counts correlate well with the volume percent water values obtained during the initial borehole soil sample analysis (see Figs. 9 and 10). The results of the volume percent moisture measurements with depth are shown in Table 4. Particle-size analysis was performed on these samples and a correlation was calculated between the uniform soil classification and moisture content. The results from repeated neutron log measurements of borehole B-17 are shown in Figure 11.

Table 4. Volume ratio of water content in soil from borehole B-17.

$\frac{\text{Vol } H_2O}{\text{Vol soil}}$	Dry bulk density	Depth (ft)	Lithology
0.370	1.870	11.000	Sandy silt
0.241	1.540	16.000	Clayey gravel
0.214	1.745	21.000	Silty sand
0.359	1.870	26.500	Silty sand
0.285	1.910	31.000	Silty sand
0.342	1.917	36.000	Sandy clay
0.337	1.860	39.000	Sandy clay

The moisture content appeared to change little during the summer of 1991. Only near the top of the borehole is there a change in moisture content. This may be due to air drying because of air leakage from the membrane. (As previously noted, this is being corrected.)

Also, the neutron log results from the SEAMIST borehole show more responsiveness (Fig. 12) than those from two boreholes (B-18 and B-19 on Fig. 1) that were completed with black iron pipe. The SEAMIST membrane attenuates the neutrons much less than does the iron pipe. Boreholes B-18 and B-19, 10 ft south of borehole B-17, were bored with the same drill as that used for borehole B-17. The results in all three boreholes seem to follow the lithologic description for the soil type; for example, soil called sand has a low moisture content and sandy clay has a higher moisture content.

FUTURE PLANS

We plan to monitor boreholes B-16 and B-17 by taking samples at 3-month intervals. The samples taken during borehole construction are being analyzed for particle size and soil classification. This will provide correlations of moisture contents in addition to the geologist's description of the lithology. We will continue to monitor boreholes B-17, B-18, and B-19 with a neutron logging device to determine any moisture movement between the wet and dry seasons in Livermore.

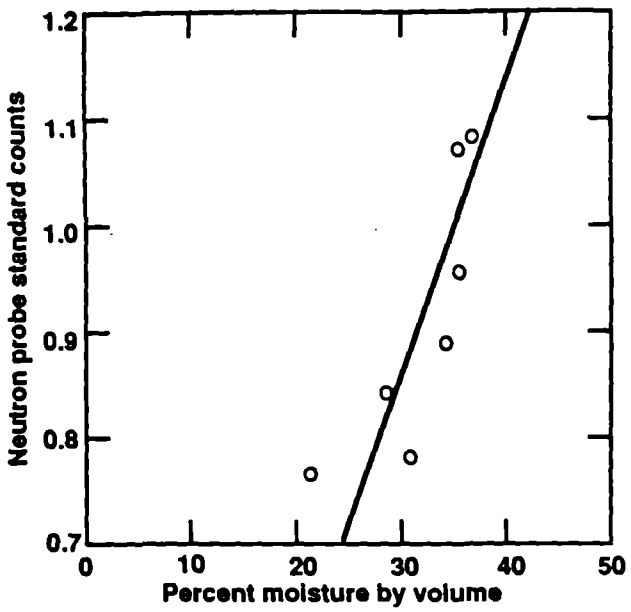

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Figure 9. Neutron probe standard counts vs volume percent water.

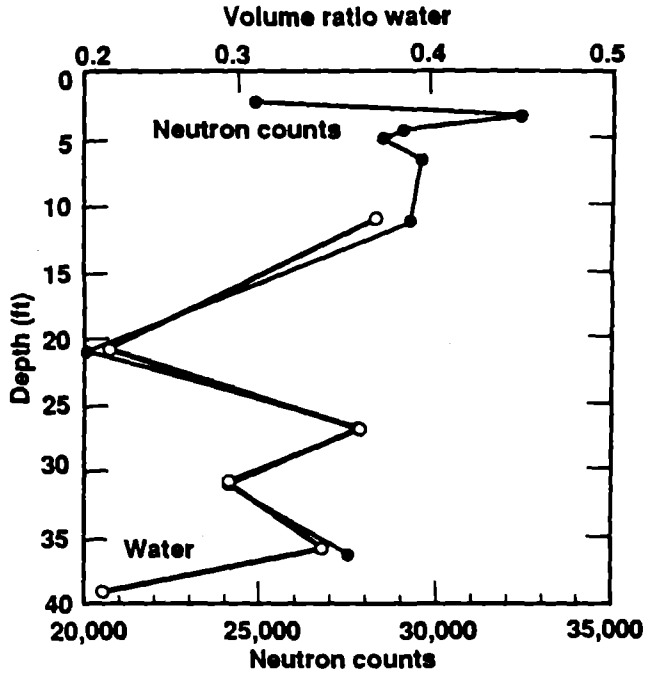

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Figure 10. Volume of water-to-soil ratio compared to neutron counts with depth in borehole UMB-292-01.

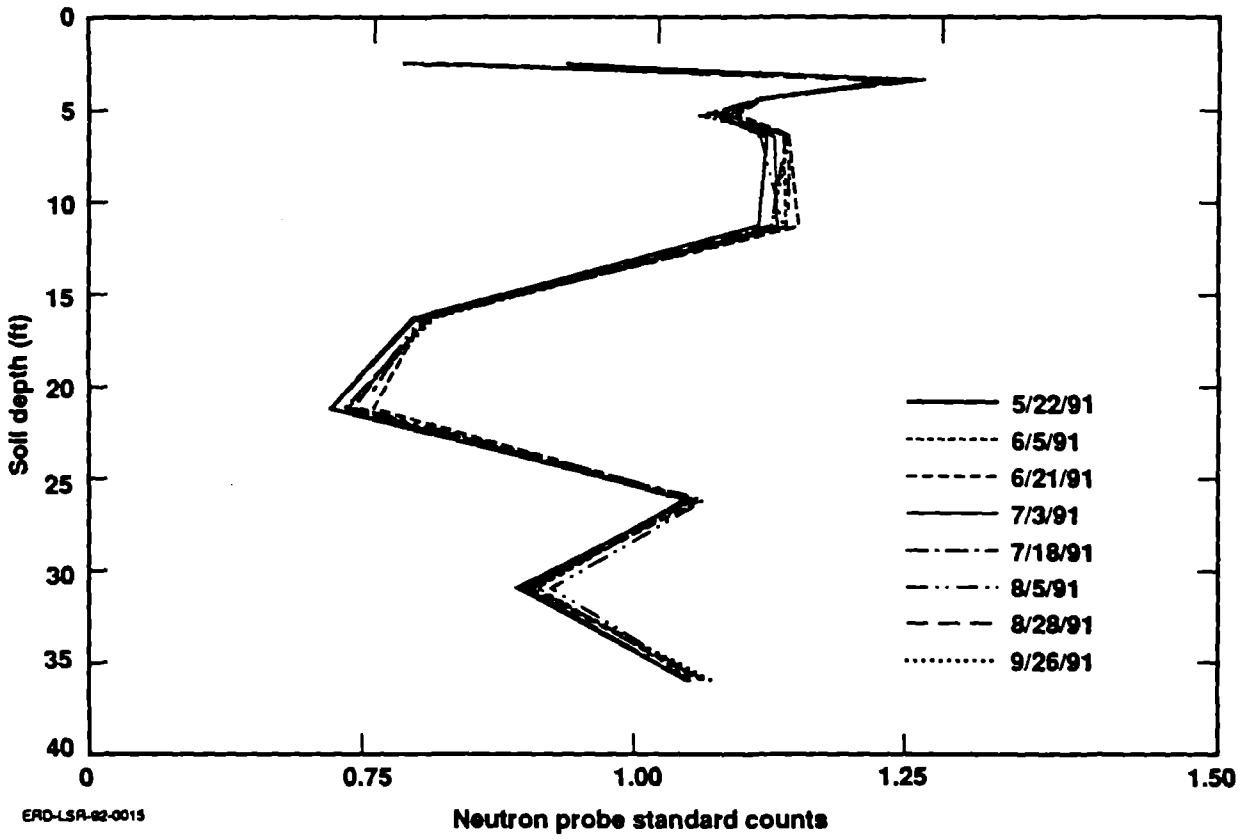

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Figure 11. Reproducibility of neutron probe standard counts vs soil depth.

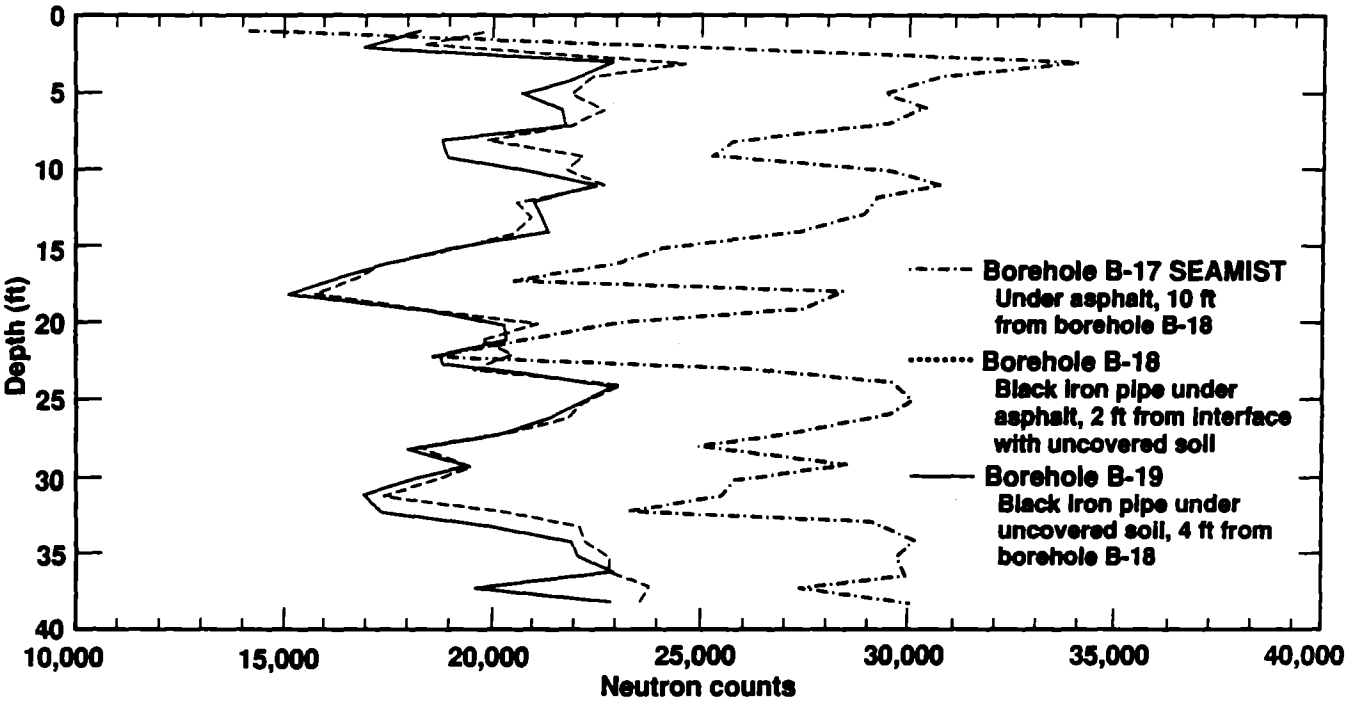

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Figure 12. Responsiveness of neutron counts in boreholes finished with black iron pipe vs those in borehole finished with SEAMIST blank membrane.

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