BUSTED BUTTE

Interim Report

PROJECT TITLE:
Radionuclide Migration Experiments in Non-welded Tuff under

Saturated and Unsaturated Conditions

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1. INTRODUCTION

An understanding of the transport of radionuclides through tuff under unsaturated and saturated conditions is essential to assessing the environmental impact of the disposal of high-level nuclear wastes in Yucca Mountain. Although field migration experiments have been performed under unsaturated conditions in non-welded tuff at the Busted Butte Unsaturated Zone Facility (BBUZF) at the Nevada Test Site, only chemical analogs of radionuclides could be used in these experiments. The transport behavior of radionuclides under these conditions must therefore be inferred from results obtained in these field experiments and a comparison between the sorptive behavior of chemical analogs and radionuclides.

This semi-annual report describes the work performed at the Whiteshell Laboratories on behalf of the United States Department of Energy, Yucca Mountain Site Characterization Office (USDOE/YMSCO), during the period November 2000 through April 2001. Results of static batch sorption results for a suite of representative tracers and radionuclides are presented. The chemical composition of the in situ pore water in the tuff is inferred from following the chemical composition of the water eluted from a trial block. Migration results for two key radioelements, Tc and Np, are presented.

1.1 Test Objectives

The objectives of this program are to study the migration behaviour of selected radionuclides through representative samples of tuff under unsaturated and saturated conditions at a scale of up to 1 m. These experiments are intended to provide experimental evidence on the transport of these radionuclides, to compare their behaviour with that observed for non-radioactive tracers performed in situ at the BBUZF, and with that predicted on the basis of laboratory sorption data obtained at the Los Alamos National Laboratory (LANL) and in supporting static batch sorption experiments at Whiteshell.

The information provided by these migration experiments is intended to contribute to the environmental assessment of Yucca Mountain as a potential used fuel repository.

1.2 Scope of Experimental Program

The work consists of the following eight related tasks:

Task 1: Develop a technique to excavate blocks of non-welded Calico Hills tuff from the BBUZF. This development will be done at Whiteshell Laboratories (WL). Because of the friable nature of excavated non-welded tuff, procedures need to be developed to prevent any excavated Calico Hills tuff from structurally disintegrating.

Task 2: Supervise the excavation of two ~1 m^3 blocks of non-welded Calico Hills tuff from the BBUZF and ship the excavated blocks to WL. Because of the fragility of the excavated non-welded tuff, it is not advisable to ship the excavated blocks by commercial transport and, consequently, the blocks may have to be transported by WL staff.

Task 3: Establish groundwater flow through the trial block under unsaturated conditions and perform a migration experiment through this block using ^{3}H, ^{22}Na, ^{60}Co, $^{95m+99}Tc$, ^{137}Cs, and ^{237}Np as well as a non-reactive, chemical tracer, Na-fluorescein.

Task 4: Using the trial block, develop a technique to obtain information on the spatial distribution of the sorbed radioisotopes, either by "fixing" sorbed tracers to the tuff so that the block can be sectioned without loss of information or by physically removing samples of the tuff.

Task 5: Establish groundwater flow and tracer migration through one of the large blocks under unsaturated conditions by applying the synthetic groundwater containing ^{3}H, ^{22}Na, ^{60}Co, ^{99}Tc, ^{137}Cs, and ^{237}Np, as well as suitable stable tracers to one or more locations at the top of the block and withdrawing the groundwater from multiple points at the bottom. For the saturated flow experiment, the same tracers will be used, but the flow direction may be horizontal rather than vertical, pending discussions with US DOE and LANL.

Task 6: Section the large blocks to obtain information on the spatial distribution of the sorbed radioisotopes using the technique developed with the smaller block.

Task 7: Determine sorption coefficients for the tracers used in the migration experiments.

Task 8: Perform chemical and radiochemical analyses by staff in the Analytical Science Branch (ASB) at WL using established and documented procedures.

2. MIGRATION EXPERIMENTS UNDER UNSATURATED CONDITIONS

2.1 Trial Block

A trial block with approximate dimensions of 38 x 38 x 30 cm was excavated from the upper zone of the Calico Hills non-welded tuff formation in the BBUZF during March and April 1999 and transported to the Whiteshell Laboratories in April. This block was used to test various techniques, to obtain preliminary data for use in Site Recommendation, and to test post-migration experiment radiometric techniques to obtain information on the spatial distribution of the radionuclides that had not been eluted at the termination of the migration experiment.

A schematic of the experiment is shown in Figure 1 and the actual experiment in progress in Figure 2.

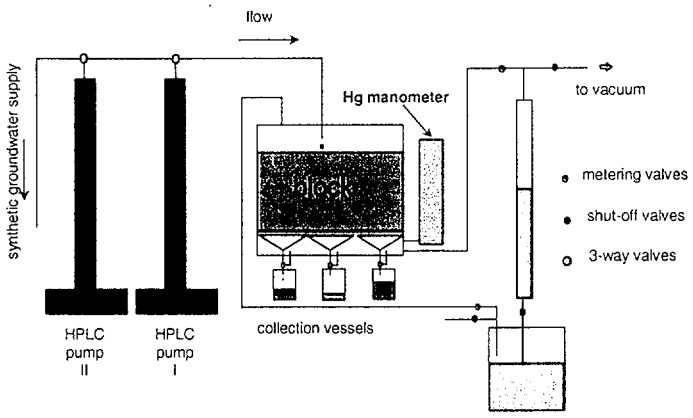

Figure 1: Schematic of Migration Experiment Under Unsaturated Conditions

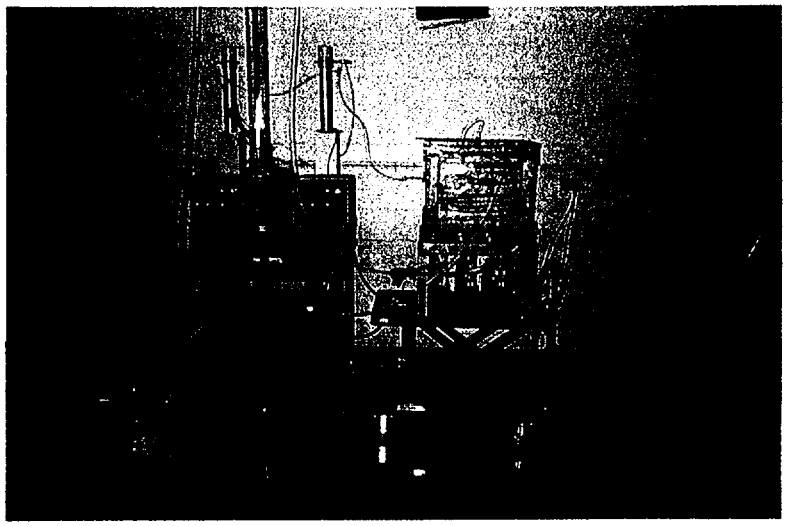

Figure 2: Migration Experiment Under Unsaturated Conditions

After a vertical unsaturated flow had been established through the block, an 800-mL volume of synthetic Busted Butte pore water containing the conservative (i.e., non reactive) tracers Na-fluorescein and $^{3}H_2O$ and the chemically reactive radionuclides ^{22}Na, ^{60}Co, $^{95m+99}Tc$, ^{137}Cs, and ^{237}Np was added drop wise to the top of the block at a rate of 20 mL/hr. This addition of the radionuclides and tracers was followed by adding synthetic Busted Butte pore water in the same way and at the same flow rate. The duration of the migration experiment was 87 days. At this time, the Np elution peak had been observed and, based on the results from static batch sorption studies (see Section 6, below), it was concluded that elution of the other radionuclides would take months to years and that it would be more economical to determine the transport of these radionuclides by determining their spatial distribution in the block.

Water exiting the trial block at the bottom was collected in a 3 x 3 array of collection bottles. These samples were analyzed spectrophotometrically for fluorescein anion concentrations, a dye tracer, and radiometrically for radionuclides.

The concentrations of the tracers eluted from the trial block are shown in Figure 3 and the recovery of the tracers that were eluted, in Figure 4. Recovery of ^{95m}Tc was about 70%. Presumably, the fraction that was not recovered has diffused away from the flow paths. Recovery of the injected $^{3}H_2O$ and Na-fluorescein approached 80%. The recovery of Np is about 0.1% but, as indicated in Figure 4, was still increasing when the experiment was terminated.

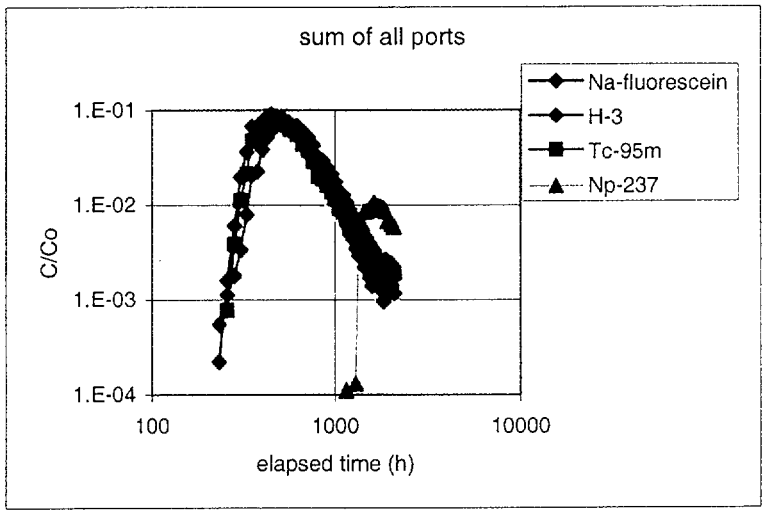

Figure 3: Elution Profiles for Na-fluorescein, TcO_4^-, NpO_2^+, and $^{3}H_2O$ from all Sample Ports Plotted as a Log-log Plot

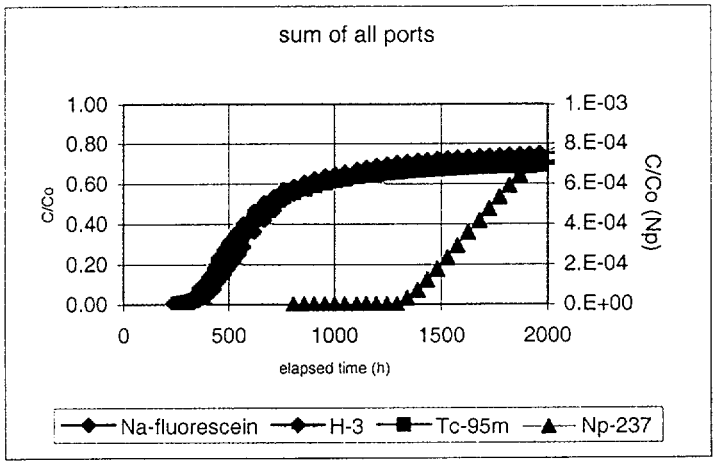

Figure 4: Recovery of Injected Tracers from the Trial Block as a Function of Time

The arrival times of the peak of the various tracers and their calculated retardation, relative to ^3H_2O, are shown in Table 1

Table 1

Transit Times and Retardation Factors

Tracer	Transit Time (h)	Retardation (^3H_2O =1)
Na-fluorescein	447	0.86
$^{95m+99}$Tc	447	0.86
^3H_2O	522	1.0
^{237}Np	1680	3.2

Rewriting the standard retardation equation,

$$R_{Np} = \frac{V_W}{V_{Np}} = 1 + \frac{\rho(1-\varepsilon)}{\varepsilon} R_{d(Np)}$$

where

V_W = velocity of conservative tracer, in this case, ^3H_2O
V_{Np} = velocity of Np
ρ = density of tuff, in this case 2.6
ε = porosity, in this case 0.5
R_d = sorption coefficient

to

$$R_{d(Np)} = \frac{\varepsilon(R_{Np} - 1)}{\rho(1-\varepsilon)}$$

a sorption coefficient of 0.85 mL/g was obtained for ^{237}Np, assuming saturated flow. For unsaturated flow, the calculated R_d value is expected to be somewhat lower. The calculated sorption coefficient falls within the range of the experimentally determined static sorption coefficients for ^{237}Np of 0.73 ± 0.34 mL/g and 3.66 ± 6.17 mL/g from solutions containing a single tracer and a mixture of tracers.

2.2. Large Block

The ~1 m^3 block, earmarked for migration experiments, was excavated from the BBUZF during October – December 1999 and transported to Whiteshell in December 1999. This block was prepared for migration experiments under unsaturated conditions using the same methodology as the trial block. The arrangement shown in Figure 1 also applies to this large block. Instead of a 3 x 3 array of collection bottles, a 6 x 6 array is used. The experimental arrangement of the large block is identical to that shown in Figure 1. The actual experiment is shown in Figure 5.

Figure 5: Migration Experiment Under Unsaturated Conditions: Large Block

In consultation with US DOE staff, two parallel migration experiments are being performed in this tuff block by adding the synthetic Busted Butte pore water and the radionuclides to two locations on the top surface of the block. However, in contrast to the method used in the trial block, where an 800- mL "top hat" injection had been used, this experiment design calls for the continuous addition of the radionuclides. The solutions are being added at a rate of 10 mL/hr to each of the two locations. The intent is to suspend the addition of the tracer solution at one of the location for an, as yet unspecified, time to look for diffusion effects.

Addition of the pore water at the two locations on the upper surface of the block was started on October 16, 2000 and the first indication of water exiting the block at the bottom was observed on March 29, 2001. By April 16, 2001, the flow through the block had stabilized sufficiently to start the addition of the tracer solutions. As of the end of April 2001, no tracers had yet been observed in the eluted pore water.

3. MIGRATION EXPERIMENTS UNDER SATURATED CONDITIONS

The purpose of this migration experiment is to study the transport behavior of radionuclides under saturated conditions as exist below the repository horizon underneath Yucca Mountain. The block of tuff excavated from the Calico Hills formation at the BBUZF has been selected for this experiment. In contrast to the vertical flow in the unsaturated zone, flow in the saturated zone is expected to be horizontal. This flow direction is reflected in the arrangement for this experiment as shown in Figure 6.

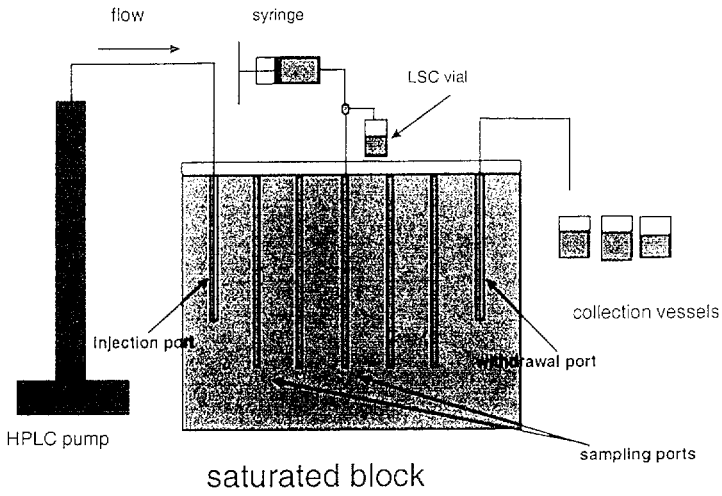

Figure 6: Schematic of Flow Under Saturated Conditions

The plywood-and-epoxy enclosure that had been built around the block prior to its excavation from the BBUZF was retained. To prevent leaks in the enclosure, a stainless steel enclosure was fabricated to fit snugly around the plywood enclosure and welded to the mild steel plate underneath the block. Two vertical, ¾" boreholes were drilled into the block to a depth of 50 cm to serve as inlet and outlet ports and to accommodate inserts to minimize dead volumes in the boreholes. Five vertical, ¾" boreholes were drilled to a depth of 75 cm between the inlet and outlet boreholes and slightly offset from a direct line between the inlet and outlet boreholes. These boreholes were equipped with inserts containing multiple sampling ports to allow small samples of solution to be withdrawn with a syringe from depths of 30, 40, 50, 60, and 70 cm during the migration experiment. Each insert consists of five lengths of Teflon tubing inserted into a polyethylene tube, with the ends exiting from the tube at 10-cm intervals. The free space inside the polyethylene tube is filled with epoxy. Finally, four, 1" boreholes were drilled near the four corners of the block to add synthetic Busted Butte pore water to saturate the block. Saturation of the block was completed by mid-February when the water level in the four boreholes reached the top of the block. At that point, loose tuff retained from drilling the various boreholes was poured into the four 1" boreholes. The upper surface of the saturated block is shown in Figure 7; this figure also shows the locations of the inlet and outlet ports and the sampling ports.

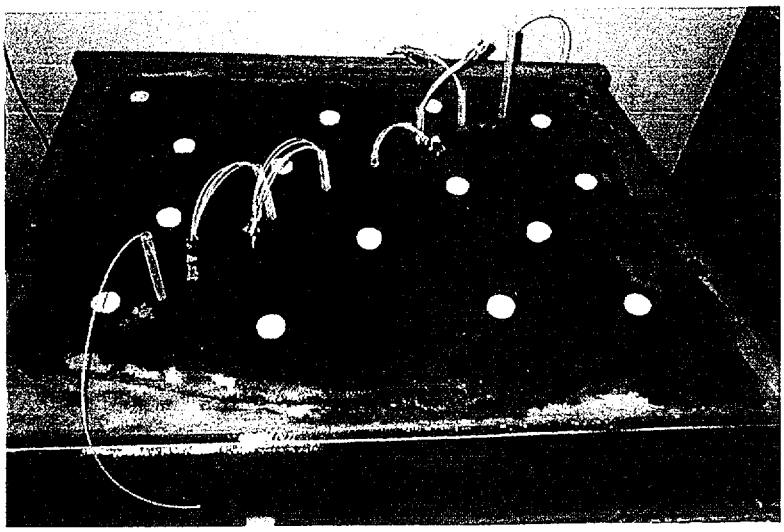

Figure 7: Upper Surface of Water-saturated Lower Block

An acrylic cover was secured to the top of the stainless steel enclosure to form a hermetic seal and synthetic Busted Butte pore water was injected into the block via the injection port using a HPLC pump at a rate of 20 mL/h during the week and 10 mL/h on weekends. On March 9, 2001, the first few drops of water were observed at the outlet port and eluted from the block. At this point, the flow from the pump was reduced to 10 mL/hr. To monitor the redox conditions in the block, a Cole-Parmer Pt electrode was installed in the Teflon line leading from the withdrawal port to the collection vessel. It is recognized that water in the block is poorly poised as the concentrations of redox couples are low and that Eh measurements may only indicate qualitative trends and cannot be interpreted as equilibrium values. However, the output voltage of the Pt electrode has started to drop and the most recent readings are in the – 300 mV range. Even after correcting for the half-cell potential (~-240 mV), the values are still negative relative to the standard hydrogen electrode.

The injection of the tracer and radionuclide solution commenced on March 27, 2001 at a flow rate of 10 mL/hr. On April 3, the first set of samples was obtained from sampling ports A and B. Sampling port A is located 10 cm downstream from the injection port and slightly off the line between the injection and withdrawal ports. The sampling ports are located 10 cm apart and in a line parallel to the line connecting the injection and withdrawal ports and are sampled weekly.

A few of the first samples retrieved from these sampling ports showed a distinct tinge of color indicating that the conservative tracer had migrated over a distance of at least 10 cm. However, the lack of color in the samples obtained from sampling port B suggests that the conservative tracer had not yet migrated over a distance greater than 20 cm over the two weeks since the injection of the tracers had been started.

To obtain some additional information on the redox condition in the saturated block, a limited amount of microbial work was performed on samples of tuff, on the synthetic pore water added to the block and on samples of water eluted from the block. The results showed that nitrate-reducing bacteria are present in the synthetic Busted Butte pore water and, quite possibly, also in the tuff. The increase in the nitrate-reducing bacteria in the water eluted from the block is

evidence of microbial activity in the block that may lead to chemically reducing conditions. This is consistent with the low Eh values obtained with the Pt electrode.

4. PORE WATER COMPOSITION IN TRIAL BLOCK

Chemical analysis of the water eluted from the trial block has shown that the target concentration of the synthetic Busted Butte pore water did not match that of the pore water in the trial block. Figure 8 shows the change in concentration of the eluted water as the experiment progressed.

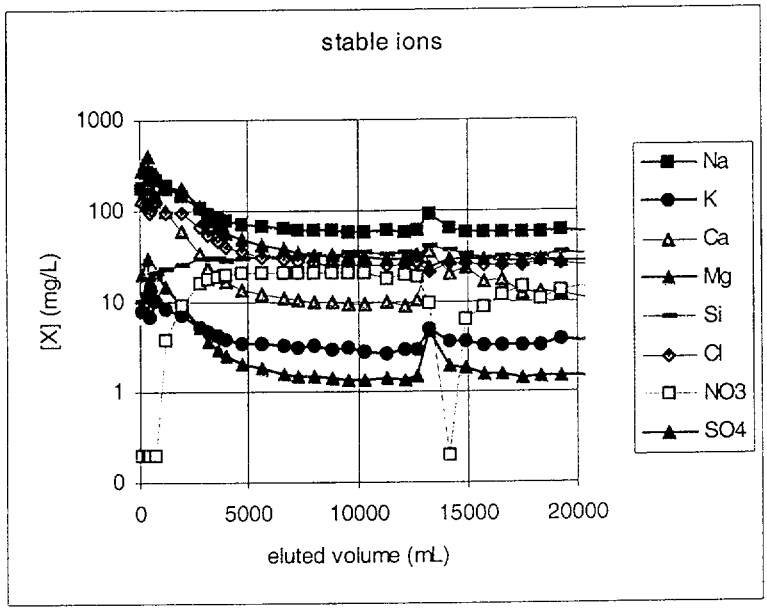

Figure 8: Cation and Anion Concentrations in Water Eluted from Trial Block

The concentrations of the cation and anions varied with time: with the exception of NO_3^-, a rather rapid initial decrease in concentration of all cations and anions was observed. This suggests that the composition of the pore fluid in the trial block was considerably different than that of the Busted Butte pore water as reported by Bussod et al. (1998). The difference in pore water composition is probably not important in the context of the migration experiment but is worth noting.

5. POST-MIGRATION EXPERIMENT RADIOMETRIC ANALYSES

Gamma scanning of the upper surface of the trial block commenced in early March 2001. A photograph of the gamma scanning system is shown in Figure 9. The gamma detector is a Princeton GammaTech IGC4020 with an efficiency of 40%; collimation is provided by a 114-mm long and 152-mm diameter 95% tungsten cylinder with a 13 mm diameter aperture.

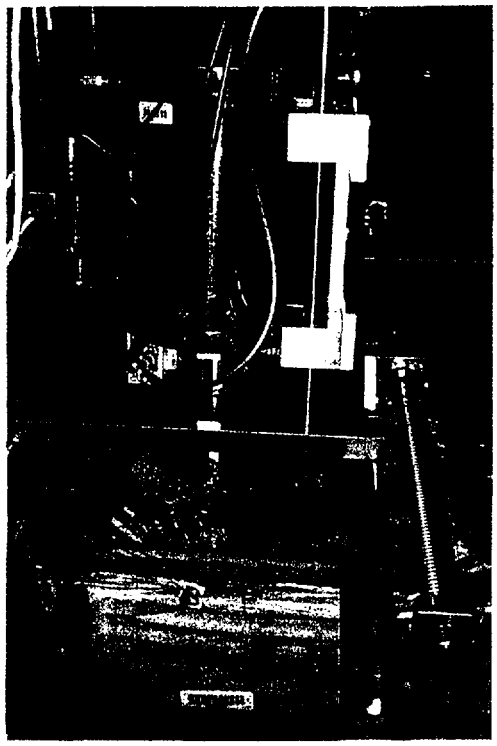

Figure 9: Ge(Li) Detector with 95% Tungsten Collimator Positioned Directly Above the Trial Block. The upper surface of the block is covered with a thin plastic film to minimize evaporative losses of moisture from the block.

Following an initial scan at a pitch of 50 mm, the upper surface of the trial block was scanned at a pitch of 13 mm, which is equal to the diameter of the collimator hole. Gamma spectra were obtained over 120 second counting periods in each location and analyzed using Genie-2000 software (Canberra Industries) and the results stored as a text file. The text file was converted into a MS Excel file and used as import data into a commercially available software plotting program, Surfer 7.02 (Golden Software, Golden, CO). This procedure generated 1,151 gamma spectra. The results were used to generate the activity distributions for ^{22}Na, ^{60}Co and ^{137}Cs using Surfer 7.02 and are plotted in Figures 10 through 12. No ^{237}Np or its daughter, ^{233}Pa, were observed above background. This is not surprising, as most of the ^{237}Np would have migrated well into the block. No ^{95m}Tc was detected either but most of this tracer was eluted from the block during the migration experiment and any remainder has by now decayed. A few points are worth noting. The physical layout of the gamma scanner restricts the diameter of the collimator to 152 mm. In spite of using 95% tungsten, which has a density of ~19 g/cm^3, the ~70-mm thickness is not sufficient to shield all gamma rays from ^{22}Na (1274 kev) and from ^{60}Co (1172 and 1332 kev). Thus, the images in Figures 10 and 11 indicate a broader spreading of the ^{22}Na and ^{60}Co than is actually the case. However, considering that the energies of the gamma rays of ^{22}Na and ^{60}Co are similar, a comparison between Figures 10 and 11 clearly shows that ^{22}Na traveled further than ^{60}Co.

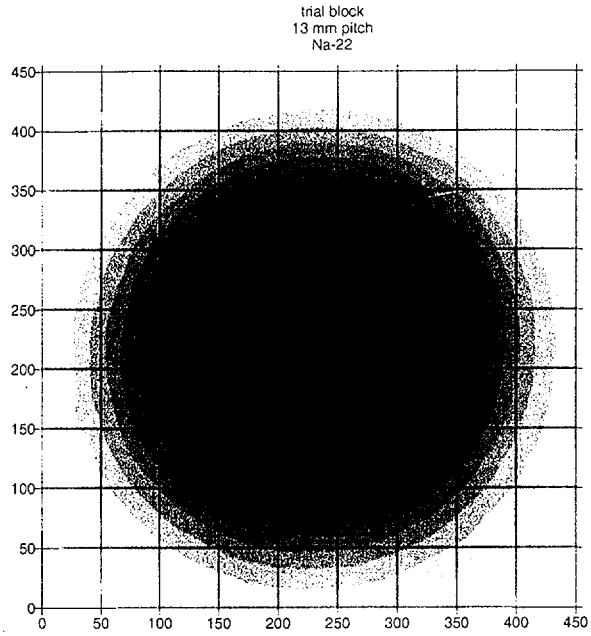

Figure 10: Spatial Distribution of ^{22}Na as Obtained by 2-D Gamma Scan of Upper Surface of the Trial Block

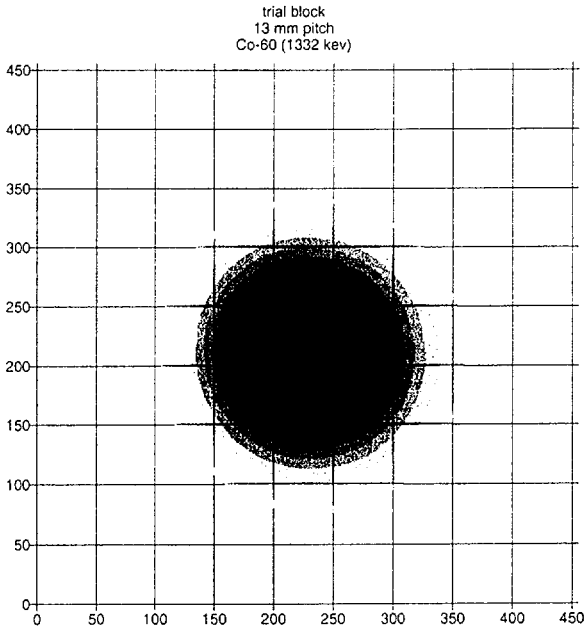

Figure 11: Spatial Distribution of ^{60}Co as Obtained by 2-D Gamma Scan of Upper Surface of the Trial Block

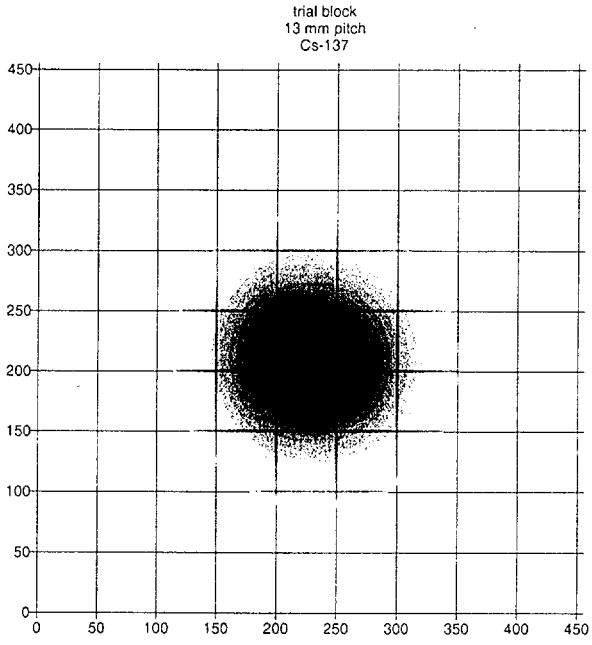

Figure 12: Spatial Distribution of ^{137}Cs as Obtained by 2-D Gamma Scan of Upper Surface of the Trial Block

Following the initial gamma scanning of the upper surface of the trial block, the block was covered with an acrylic plate with a square opening in the center and with reference marks at 1 cm intervals. Using a length of plastic tube with a square, ~1 cm x ~1 cm cross section, a thin, ~ 2 mm, layer of material was aspirated into a tared polyethylene counting vial and weighed and submitted for radiometric analysis. A total of 81 samples were obtained from the center of the upper surface in a 9 x 9 cm grid. Figure 13 shows the removal in progress and Figure 14 shows the upper surface of the trial block after the first 9 samples had been removed. Contour plots were generated using Surfer 7.02 and are plotted for the three main radioisotopes in Figures 15 through 17. Note the amounts of activity as indicated in the color bars along the side of each contour plot. The ^{60}Co is concentrated near the center, suggesting that movement of this radioisotope was very limited. The contour plot for ^{137}Cs shows lower concentrations near the center, while the contour plot for ^{22}Na shows that the radioisotope is virtually depleted from the center. This is qualitatively consistent with the observed static batch sorption data. Summing the amounts of activity detected, 0.04%, 6.4%, and 4.3% of the ^{22}Na, ^{60}Co, and ^{137}Cs were removed with the first few mm of the tuff. Again, this is qualitatively consistent with the static batch sorption coefficients for these radioisotopes, which predicts that the most strongly sorbing tracer, ^{60}Co, should be present in the highest concentration near the inlet. Exact values for the depth of excavation are not yet available, but will be calculated from the weight of the excavated samples after drying.

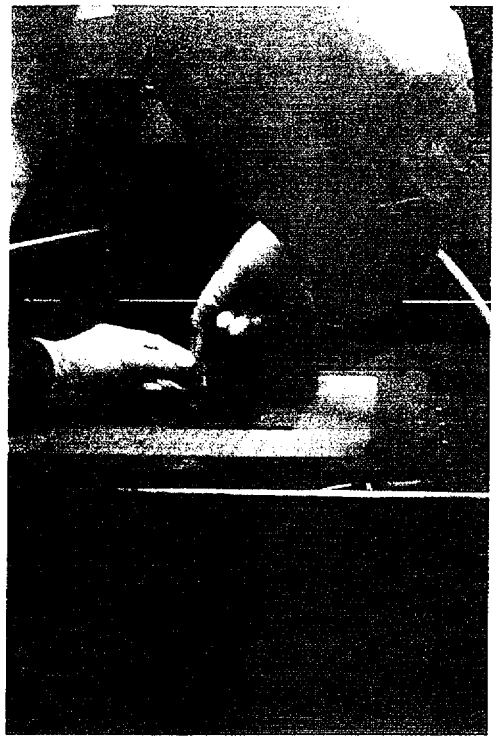

Figure 13: Removal (by aspiration) of Loose Material from an Isolated Area of the Upper Surface of the Trial Block

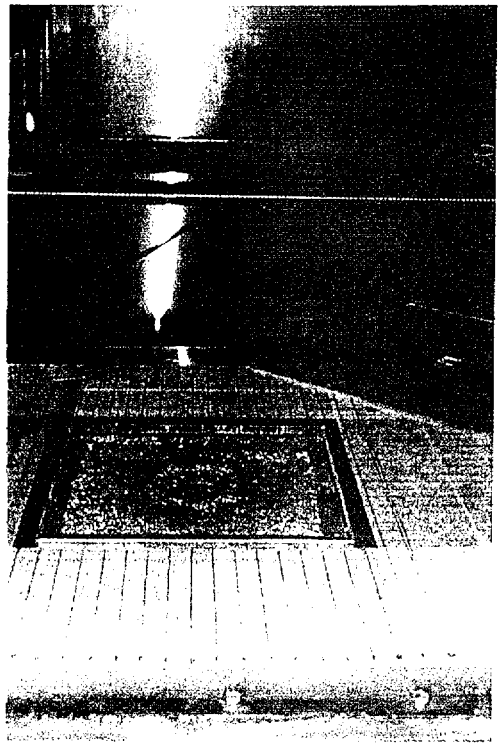

Figure 14: Upper Surface of the Trial Block after Removal of the First 9 Samples in a 3 cm x 3 cm Grid

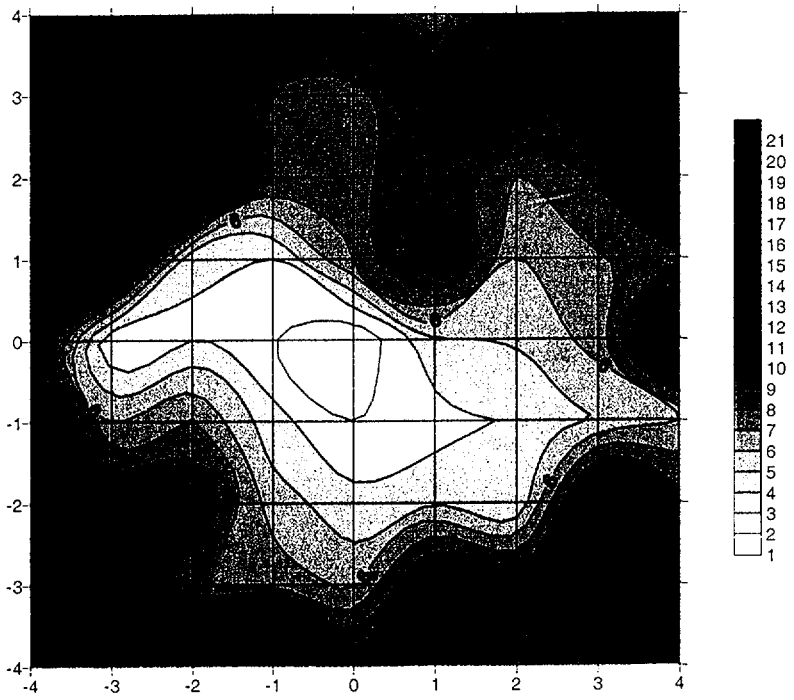

Figure 15: Contour Plot for ^{22}Na Obtained on Material Removed from a 9 x 9 cm Area at the Top of the Trial Block. Units in Bq/sample.

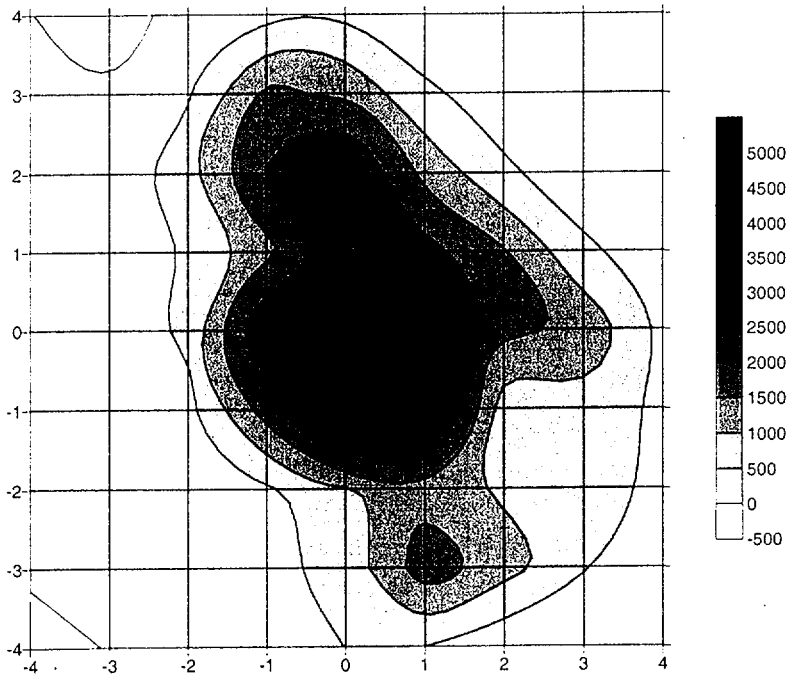

Figure 16: Contour Plot for ^{60}Co Obtained on Material Removed from a 9 x 9 cm Area at the Top of the Trial Block. Units in Bq/sample.

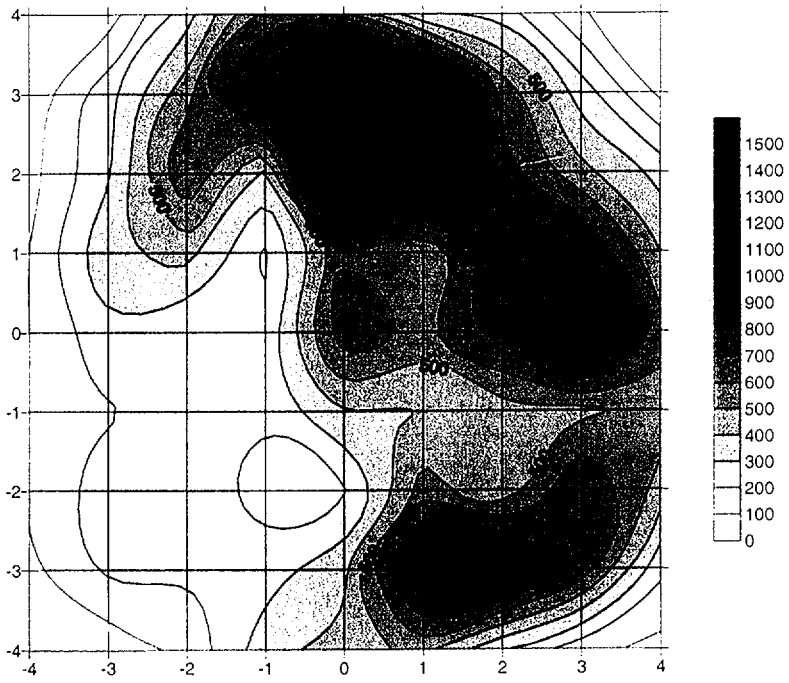

Figure 17: Contour Plot for ^{137}Cs Obtained on Material Removed from a 9 x 9 cm Area at the top of the Trial Block. Units in Bq/sample.

After the first 81 samples were removed, additional material was removed from the same locations but to a depth of ~5 mm. These samples have also been submitted for radiometric analysis but results were not yet available by the end of April.

6. MICROBIAL INVESTIGATIONS

The results of microbiological analyses of the synthetic Busted Butte pore water prepared at Whiteshell, of the water being eluted from the block, and of samples of tuff, obtained from the upper and lower tuff blocks using a sterile drill and storing the samples in a sterilized container, indicate that there is some microbial activity in the block. The results of these analyses are shown in Table 2.

Dr. Sim Stroes-Gascoyne, who supervised these investigations at the Whiteshell Laboratories, provided the following background information and discussion of the results from the microbial investigations.

Nitrate-reducing bacteria (NRB) are often facultative organisms, which use aerobic respiration with O_2 as electron acceptor in the presence of air (even if nitrate is present) but use nitrate as electron acceptor in the absence of O_2. Nitrate-reducing bacteria reduce nitrate, in dissimilatory metabolism using a number of enzymes, via nitrite, nitric oxide and nitrous oxide, to dinitrogen (N_2). Some can reduce nitrate to NH_4^+ in dissimilative metabolism. Many NRB will also reduce other electron acceptors anaerobically such as ferric iron, and certain organic electron

acceptors, and many can grow by fermentation. Therefore, NRB are metabolically quite diverse in terms of alternative energy-generating mechanisms but nitrate reduction yields less energy than aerobic respiration (Madigan et al. 2000).

Table 2 shows the enumeration results for all samples, i.e., aerobic and anaerobic heterotrophs obtained on two media, R2A and R2A + nitrate. The heterotroph concentrations are expressed as Colony Forming Units (CFU) per unit mass or volume. Table 2 also contains the data obtained for NRB and SRB (sulfate-reducing bacteria); these are expressed in terms of Most Probable Number (MPN) per unit mass or volume. The presence of nitrate is not expected to make a difference to the amount of viable aerobes that can be cultured under aerobic conditions. The R2A medium is not N-limited (for assimilatory processes, i.e., the building of cells) and dissimilatory nitrate reduction (for energy production) occurs only under anaerobic conditions. Therefore, aerobic enumeration on R2A with and without nitrate added should give identical results. This is indeed the case for all samples enumerated in Table 2, except for one sample from the saturated block, in which there the results from the two media vary by less than a factor of two. This close agreement between the results obtained from the two media for all aerobically enumerated samples is a good indication of the accuracy of the methods used and provides an extra degree of confidence in the results.

For anaerobically analyzed samples, it is expected that, if NRB are present (i.e., viable and culturable), R2A + nitrate medium would give higher results than R2A. Table 2 shows that in most cases (i.e. all the rock samples), anaerobic results on R2A + nitrate are in fact lower than on R2A, suggesting that none of these rock samples contained large amounts of viable and culturable NRB. The fact that SRB are not present either and that generally anaerobic heterotrophic enumeration results are low compared to aerobic results suggests that these rock samples came from very aerobic environments with either no NRB population or, if NRB were present, they were not adapted to anaerobic conditions and nitrate reduction for energy.

Nitrate-reducing bacteria were also enumerated in a more specific manner. Results (Table 2) suggest that NRB were present only in liquid samples, i.e. 1.1×10^2 MPN/mL inflow and $>10^4$ to 5×10^6 MNP/mL outflow. Anaerobic enumerations on R2A and R2A + nitrate have not been counted yet for the outflow samples, so a comparison cannot be made except for the inflow sample where anaerobic heterotrophs on R2A with nitrate yielded somewhat higher results than in most rock samples. Therefore, although the rock samples did not contain significant amounts of viable and culturable NRB, the inflow sample did contain them, and the outflow samples contained large populations of NRB. This suggests that an anaerobic environment was created in the saturated block that induced the proliferation of NRB. Their presence explains the reduction in nitrate levels in the outflow compared to the inflow water nitrate concentrations.

Table 2

Microbial Analysis Results

Block	Sample and sample treatment	Aerobic Heterotrophs CFU/g dry wt or mL water	Anaerobic Heterotrophs CFU/g dry wt or mL water	NRB MPN/g dry wt or mL water	SRB MPN/g dry wt or mL water
Saturated Block	Synthetic pore water for Saturated Block	$(2.23 \pm .14) \times 10^5$ R2A $(2.29 \pm .2) \times 10^5$ R2A + N	$(2.86 \pm .23) \times 10^2$ R2A $(7.13 \pm .4) \times 10^1$ R2A + N	110 UL = 470 LL = 76	<0.41
Saturated Block	Ground tuff from inlet port	$(5.29 \pm .64) \times 10^4$ R2A $(5.48 \pm .84) \times 10^4$ R2A + N	$(3.86 \pm 6.72) \times 10^0$ R2A $(3.86 \pm 6.69) \times 10^0$ R2A + N	<3.48	<4.7
Saturated Block	Ground tuff from outlet port	$(1.19 \pm .03) \times 10^6$ R2A $(1.16 \pm .02) \times 10^6$ R2A + N	$(1.29 \pm .4) \times 10^2$ R2A $(3.58 \pm 6.2) \times 10^0$ R2A + N	< 3.22	<4.4
Saturated Block	Ground tuff from sample port	$(1.95 \pm .04) \times 10^6$ R2A $(2.04 \pm .1) \times 10^6$ R2A + N	$(4.85 \pm 2.40) \times 10^1$ R2A $(4.57 \pm .45) \times 10^2$ R2A + N	< 3.12	<4.2
Saturated Block	Water eluted from saturated block; sample E-0-5	NA	NA	4.6×10^6 UL = 2.1×10^7 LL = 1.0×10^6	>11
Saturated Block	Water eluted from saturated block; sample E-0-9	$(3.67 \pm .19) \times 10^6$ R2A $(1.99 \pm .25) \times 10^6$ R2A + N	Not counted yet	$>1.1 \times 10^4$	0.41 UL = 2.9 LL = .056
Unsaturated Block	Ground tuff from top sampling port	$(3.27 \pm 1.09) \times 10^2$ R2A $(5.09 \pm 1.09) \times 10^2$ R2A + N	<1.0 R2A <1.0 R2A + N	<3.27	<4.4
Unsaturated Block	Ground tuff from middle sampling port	$(4.98 \pm .54) \times 10^3$ R2A $(4.33 \pm 2.17) \times 10^3$ R2A + N	$(3.98 \pm 3.25) \times 10^1$ R2A $(3.58 \pm 10.8) \times 10^0$ R2A + N	<3.25	<4.4
Unsaturated Block	Ground tuff from bottom sampling port	$(1.51 \pm .25) \times 10^5$ R2A $(1.54 \pm .12) \times 10^5$ R2A + N	< 1.0 R2A $(7.68 \pm 11.5) \times 10^0$ R2A + N	< 3.4	<4.7

CFU = Colony Forming Units
MPN = Most Probably Numbers
UL = Upper Limit
LL = Lower Limit
NRB = nitrate reducing bacteria
SRB = sulfate reducing bacteria

7. SUPPORTING STATIC BATCH SORPTION STUDIES

Static batch sorption experiments were performed in support of the radionuclide migration experiments using the method by Weaver (1996). These experiments were carried out in triplicate for each of two types of geological material (Topopah Spring Tuff, crystal-poor, vitric zone 1, and Calico Hills Formation) and two synthetic groundwaters (synthetic Busted Butte

pore water and synthetic J-13 groundwater) at two liquid volume to mass ratios (nominally 5:1 and 20:1) for five individual radioactive tracers (^{22}Na, ^{60}Co, $^{95m,99}Tc$, ^{137}Cs and ^{237}Np) and for Na-fluorescein as well as solutions containing a mixture of all radionuclide and dye tracers. In all cases, triplicate blanks (centrifuge tubes containing the tracer solution but no geological material) were used to determine the extent of tracer interaction with the reaction vessel.

Samples of material from the two geological formations were disaggregated by hand. Approximately 1-gram portions of the material were weighed into tared 30-mL polycarbonate centrifuge tubes, which were used as the reaction vessels. The geological materials were conditioned for a period of 7 d with synthetic pore water or groundwater prior to initiating the sorption step. The conditioning solution was removed following centrifugation, and the amount retained was determined by weight. The appropriate tracer solution was added and the sorption reaction was allowed to occur for 21 d. Following the sorption period, the separation of the solid and liquid phases was ensured by three successive centrifugations prior to removing aliquots of the aqueous phase for analysis. The radiotracer concentrations were determined by gamma spectroscopy and the Na-fluorescein concentrations using an HP 8542A Diode Array Spectrophotometer.

In general, the tracer concentrations in the blanks and the initial solutions were within the analytical uncertainty of the measurement. However, in the mixed tracer sorption experiments the concentration of Co in the blanks for the J-13 solution was found to be somewhat lower than in the initial solution.

The sorption values were calculated using the equation

$$R_d = \frac{V}{m} \times \frac{C_0 - C_f}{C_f}$$

where V is the volume of contacting solution
m is the mass of geological material
C_o is the initial tracer concentration, and
C_f is the final tracer concentration following the reaction period.

The results are shown in Table 3

Table 3

Static Bath Sorption Results

Tracer	R_d (mL/g) (single tracer)	R_d (mL/g) (mixture of tracers)
Na-fluorescein	-0.13 ± 0.26	0.45 ± 0.27
Tc	-0.06 ± 0.20	-0.06 ± 0.18
Np	0.73 ± 0.34	3.66 ± 6.17
Na	6.35 ± 0.33	2.44 ± 0.17
Cs	1360 ± 600	250 ± 0.14
Co	2050 ± 600	440 ± 60

8. SUMMARY AND PRELIMINARY CONCLUSIONS

Groundwater flow has been established in the blocks under saturated and unsaturated conditions. Migration experiments with a dye tracer and with radioisotopes were initiated in both blocks during this reporting period.

Microbial investigations have shown a considerable amount of microbial activity. This may have implications for the redox conditions in the saturated block and for the transport of multivalent radioisotopes.

Post-experiment radiometric analysis of the trial block has begun. Gamma scanning of the upper surface of the block at a pitch of 13 mm was completed and the removal of thin layers of tuff from the top of the block started. The preliminary results indicate that the transport velocity of ^{137}Cs was higher than that of ^{60}Co but lower than that of ^{22}Na. This agrees qualitatively with our observed static batch sorption coefficients.

REFERENCES

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