Characterization of the Contaminated Soil Under the
Hanford 324 Building B Cell, Washington, USA - 12182

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ABSTRACT

The 324 Building on the Hanford site played a key role in radiochemical and metallurgical research programs conducted by DOE. The B hot cell in the 324 Building was the site of high-level waste vitrification research. During cleanout operations in November 2009, a tear was noted in the stainless steel liner on the floor of B Cell. Exposure rate readings taken at various locations in the soil about 0.5 meters below B Cell reached 8,900 Roentgen (R) per hour, confirming the existance of a significant soil contamination field. The source of the radioactive material was likely a 510 L spill from the Canister Fabrication Project, consisting of purified, concentrated Cs-137 and Sr-90 solutions totaling 48,000 TBq (1.3 MCi). MCNP modeling was used to estimate that the measured exposure rates were caused by 5,900 TBq (160 kCi) of Sr-90 and Cs-137, although additional contamination was thought to exist deeper in the soil column. Two physical soil samples were obtained at different depths, which helped verify the contamination estimates. A detailed exposure rate survey inside B Cell was combined with additional MCNP modeling to estimate that an additional 1,700 TBq (460 kCi) is present just below the floor.

FRG CANISTER FABRICATION PROJECT

The 324 Building was built on the Hanford site 300 Area in 1966 as part of the Atoms for Peace program. From the outset, the B hot cell in the 324 Building was intended to house high-level waste vitrification research. Initial research focused on vitrification of first cycle waste from defense sources. Subsequent research was designed to support commercial fuel reprocessing.

In November 1984, DOE entered into an agreement to fabricate heat and radiation sources for the Federal Republic of Germany (FRG) to test fuel repository designs in the Asse salt mine. The FRG Canister Fabrication Project was initiated to fabricate the sources in the 324 B Cell, using a radioactive liquid-fed ceramic melter (RLFCM) designed and tested as part of prior research [1].

The FRG sources were intended to simulate the properties of spent reactor fuel, but without the associated actinide content and long-term degradation issues. A borosilicate glass matrix was utilized for long term stability. The glass matrix was loaded with Cs-137 to provide the required radiation and with Sr-90 to provide the required decay heat [1].

Initially two test canisters were made in run RLFCM-6. The lessons learned with the two test canisters were incorporated into the process design, and three runs of production canisters were completed. In 1987 the contract was amended to include a pair of instrumented canisters to be used to validate heat transfer models.

The canisters were subjected to a rigorous radiological and chemical characterization program to satisfy contractual requirements. Samples of glass were obtained from 2 of 3 production pours and subjected to laboratory analysis. The analytical results showed unexpected

varaibility in the Sr-90 and Cs-137 content, and an unexpectedly high concentration of actinides. Exposure rates from the completed canisters were measured inside the cell, and the thermal outputs were calculated based on the isotopic content. The characteristics of the FRG canisters are summarized by run in Table I [1].

Table I. FRG Canister Summary

Run	Canister Count	Average Properties			
		Cs-137 Activity (kCi)	Sr-90 Activity (kCi)	Decay Heat (W)	Surface Exposure Rate (kR/hr)
RLFCM-7	10	192	85	1490	272
RLFCM-8	10	78	143	1330	112
RLFCM-9	10	207	130	1860	310
RLFCM-10	2	136	59	1047	210

Ultimately, the FRG government was compelled to cancel the Canister Fabrication Project because they were unable to license their Asse salt mine repository. The canisters were cleaned, packaged in heavily shielded shipping casks, and shipped to a storage pad in the Hanford 200 W area, where they remain.

FEED SOLUTION LEAK

On October 21, 1986, during routine transfers of radioactive liquid feed material for the FRG Canister Fabrication Project, approximately 510 liters of concentrated Cs-137/Sr-90 solution leaked onto the floor of B Cell. The estimated radiochemical inventory of the solution was 33,000 TBq (883 kCi) Cs-137 and 14,000 TBq (388 kCi) Sr-90 [2].

After the spill, B Cell was extensively cleaned. However, the removed activity was far less than the known quantity spilled. The fate of the radioactive material unaccounted for was for years a source of speculation.

It is now thought that the stainless steel liner on the floor of 324 B Cell had by that time failed, and that a portion of the spilled liquids passed through breaches in the liner. Since the floor of B Cell is sloped to allow liquids to accumulate in the sump, a significant portion of the spill was able to reach the soil column. A photograph of the failed liner is shown in Figure 1.

The design of B Cell featured a 6 inch thick floor slab on compacted earth, 10 feet below grade, and completely surrounded by an asphalt-impregnated felt expansion joint. Consequently, there was little to prevent liquids penetrating the liner from reaching the soil column below B Cell.

GEOPROBE MEASUREMENTS

The tear in the B Cell liner was not discovered until November 2009, when process equipment positioned over the area and contaminated grout that had collected in the sump had been removed as part of cell decommissioning activities. At first, the tear was thought to be a recent event, caused by the pneumatic tools used for grout removal.

Since the liner had been breached, there was concern that a soil contamination field may exist under B Cell. To assess the potential for soil contamination issues, a single instrument housing was to be driven horizontally under the B Cell sump using a Geoprobe® driver. The Geoprobe

driver was located in an excavation site approximately 50 feet away from the 324 building. The instrument housing was simply a hollow steel tube, threaded together in sections, and closed on one end with a cone penetrator. The Geoprobe driver and instrument housing system allowed standard survey instrumentation to be precisely positioned along the length of the tube to obtain measurements, and then be retrieved without becoming externally contaminated.
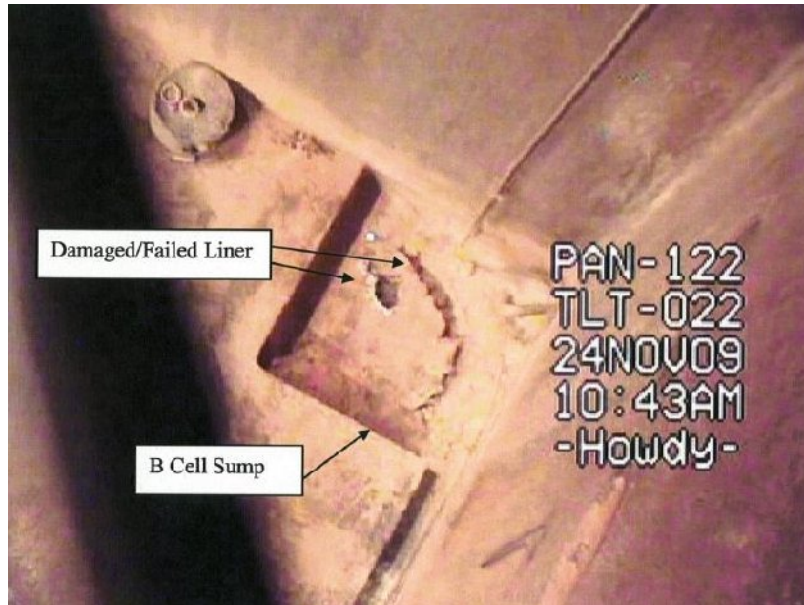

Figure 1. B Cell Liner Failure

The first instrument housing hit the cell foundation and was abandoned in place. Measurements in the second, slightly lower housing revealed a peak exposure rate of 6,700 Roentgen per hour, well in excess of the expected readings. As a result, the campaign was expanded to include 9 total housings at different angles and elevations to encompass as much of the contamination zone as possible. Subsequent measurements identified a peak exposure rate of 8,900 Roentgen per hour. The Geoprobe sampling campaign is summarized in Figure 2.
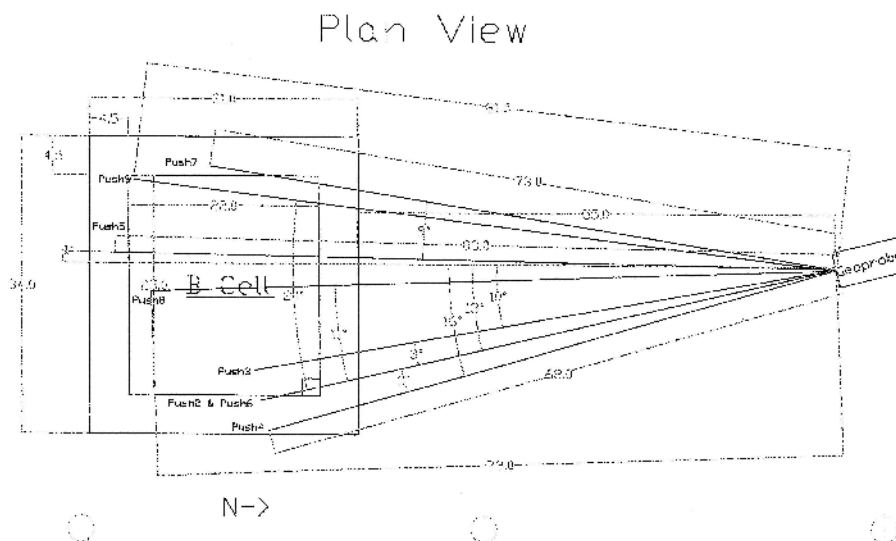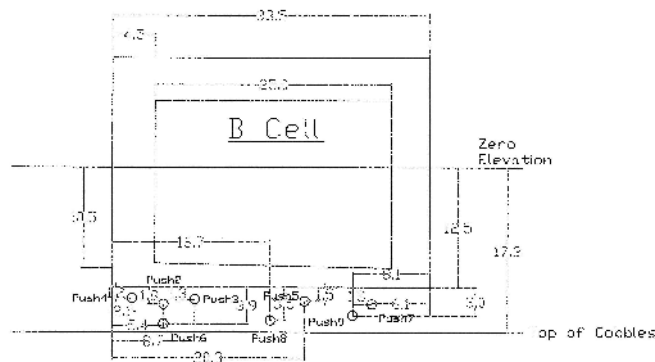
Figure 2. Geoprobe Sampling Campaign

Exposure rate readings were measured using the Eberline RO-7 system [3], which features a hand-held instrument connected to a low-range, mid-range, or high-range probe. Each probe consists of an air-filled ion chamber sized to provide the desired sensitivity, as well as a matched pre-amplifier capable of transmitting the signal through up to 500 feet of cable. Use of the RO-7 system allowed the probes to be extended down the instrument housings while the instrument remained at the excavation site.

The magnitude of the exposure rates measured during the Geoprobe sampling campaign indicated that many thousands of curies had been released to the soil. The only event with comparable quanties of radioactive material unaccounted for was a 510 L spill during the FRG Canister Fabrication Project. Consequently, the liner was assumed to have failed prior to the spill, and the radioactive material in the soil contamination field was assumed to be purified Cs-137 and Sr-90. Efforts were immediately initated to determine the total inventory in the soil contamination field, to determine the physical extent of the soil contamination field, and to verify the composition of the radioactive material in the soil.

GEOPROBE ACTIVITY CALCULATIONS

In an effort to determine the total inventory in the soil contamination field, the exposure rate measurements obtained from the Geoprobe sampling campaign were converted to activities using MNCP modeling [4]. Separate MCNP models were developed for each of the instrument housings. The instrument housings were assumed surrounded by a column of 1 cubic foot source cubes, with radioactive material uniformly distributed within the source cube but varying from cube to cube.

Photon Spectrum

The photon spectrum of the FRG Canister Fabrication Project feed material was calculated using the Oak Ridge Isotope Generation (ORIGEN2) code [5]. Separate spectra were first calculated for 1 curie of Cs-137 and Sr-90. Both spectra used the bremsstrahlung library for water. Then the two spectra were merged using the activity ratios in the spill. The merged spectrum was used to simulate 1 curie of FRG Canister Fabrication Project feed material in each source cube.

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The predominant contributor to exposure rate was Cs-137. Previous experience with ORIGEN2 spectra indicates that the 575 keV mean energy bin structure used by ORIGEN2 under-predicts the exposure rate from 662 keV photons from Cs-137 decay. The magnitude of the under-prediction is dependent on the type and thickness of shielding material between the source and measurement point, and can vary from essentially zero for air to slightly greater than 44% for 2.54 cm (1 in) of lead. Therefore, the mean energy of the 575 keV bin was shifted to 662 keV, and the photon rate for the bin was reduced to maintain the same energy rate.

Materials

The materials of construction associated with the Geoprobe models include carbon steel, Hanford soil, and air. Material compositions and densities were taken from PNNL-15870 [6] whenever possible. Since PNNL-15870 does not include a soil composition, Hanford soil was assumed to have the same composition as granite, and a density of 1.670 grams per cubic centimeter [7].

Tallies

Point detector tallies were placed inside the Geoprobes at all locations where an exposure rate had been measured. To simulate the performance of RO-7 probes, each tally utilized the FM card to calculate the ionization of air, and a tally conversion factor to yield results in R/hr.

Source distribution flagging was used for each tally to separate the contributions from each source cube to each exposure rate measurement point. This approach allowed for the construction of a matrix equation relating the unknown source cube activities to the known exposure rate measurements.

Tally Conversion Factor

MCNP tallies are reported in units of photon fluence per history, while the exposure rates measured using RO-7 probes are recorded in units of Roentgen per hour. Often MCNP calculations utilize fluence-to-dose factors to convert tally results to dose, and then assume that 0.01 Sv (1 rem) of dose is equivalent to one Roentgen of exposure. Use of this assumption introduces a bias whose magnitude is dependent on the source of the conversion factors used and the photon spectrum of the material in question.

The tally multiplier (FM) card provided in MCNP may be used to convert fluence tallies directly to Roentgen, thereby avoiding the bias introduced by using fluence-to-dose conversion factors. To assess the magnitude of the bias, a Cs-137 point source surrounded by air was modeled using MCNP. Tallies were positioned at different distances from the point source. The tally fluences were converted directly to dose rate using the DE/DF cards, and also to exposure rate using the FM card. The dose rates from several common sources of fluence-to-dose factors were compared to the exposure rate to determine bias. The bias values are shown in Table II.

Table II. Fluence to Dose Bias

Fluence-Dose Factors	Bias
ANSI/ANS-6.1.1-1977 [8]	15.6%
ICRP 21 [9]	-1.6%
ANSI/ANS-6.1.1-1991 [10]	-7.0%
ICRP 51 [11]	-7.0%

For this effort, the FM card approach was used to calculate exposure rate directly, thereby improving accuracy. The factor required by the FM card was taken from Reference [12].

$$C = 2.691 \cdot 10^{-6} \left(\frac{\text{sec}}{\text{MeV}} \right) \cdot \left(\frac{\text{cm}^2}{\text{b}} \right) \cdot \left(\frac{\text{R}}{\text{hr}} \right)$$

(Eq. 1)

Solver

The source activity to exposure rate conversion factors (F) calculated by MCNP, the unknown radioactive source activities (S), and the exposure rate measurements (ER) were cast as a matrix equation as follows.

$$F \cdot S = ER$$

(Eq. 2)

Since there were more exposure rate measurements than source activities, this equation was not square and could not simply be inverted to calculate the unknown source activities. Instead, the Conjugate Gradient solver built into Microsoft Excel® was used to calculate the activity in each source cube, and to enforce constraints that each source activity had to be greater than or equal to zero.

For each instrument housing the solver was initialized with one curie in each source cube. The solver was then executed multiple times to interatively solve for the unknown source activities. In some cases, the source activities had to be manually adjusted to help the solver converge. The final activities associated are summarized in Figure 3 and Table III.
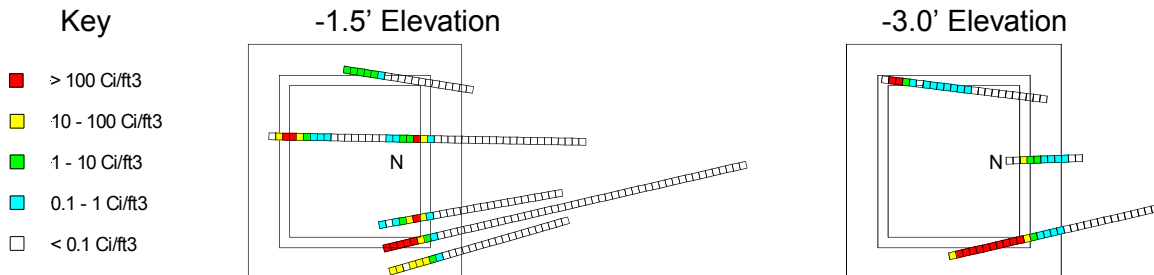

Figure 3. Geoprobe Activities

Table III. Geoprobe Activities

Instrument Housing	Cs-137 Activity (Ci)	Sr-90 Activity (Ci)	Total Activity (Ci)
Push 2	1.92E+03	8.42E+02	2.76E+03
Push 3	5.69E+02	2.50E+02	8.20E+02
Push 4	1.63E+02	7.16E+01	2.35E+02
Push 5	1.46E+03	6.43E+02	2.11E+03
Push 6	2.86E+03	1.26E+03	4.11E+03
Push 7	1.32E+01	5.78E+00	1.89E+01
Push 8	2.77E+01	1.22E+01	3.99E+01
Push 9	7.72E+02	3.39E+02	1.11E+03
Total	7.78E+03	3.42E+03	1.12E+04

CONTAMINATION FIELD ACTIVITIES

From Figure 3 it is clear that the soil contamination beneath B Cell is widespread, and features a ring of high activity soil in the vicinity of the expansion joint in the floor. The contamination also appears to spread out horizontally and reduce in magnitude as the radioactive material descended vertically. These characteristics were used to construct the simplified contamination field shown in Figure 4.

A statistical examination of the source activities indicated that there were distinct high, medium, and low activity populations, each with a log-normal distribution of source activities. Population means were developed for each by splitting the calculated activities into three populations using activity thresholds. The thresholds were set iteratively such that small changes in the thresholds would not significantly affect the calculated means.

Soil volumes derived from Figure 4 were multiplied by the mean activity concentrations to yield the total activity of the contamination field. The contamination field calculations are summarized in Table IV.
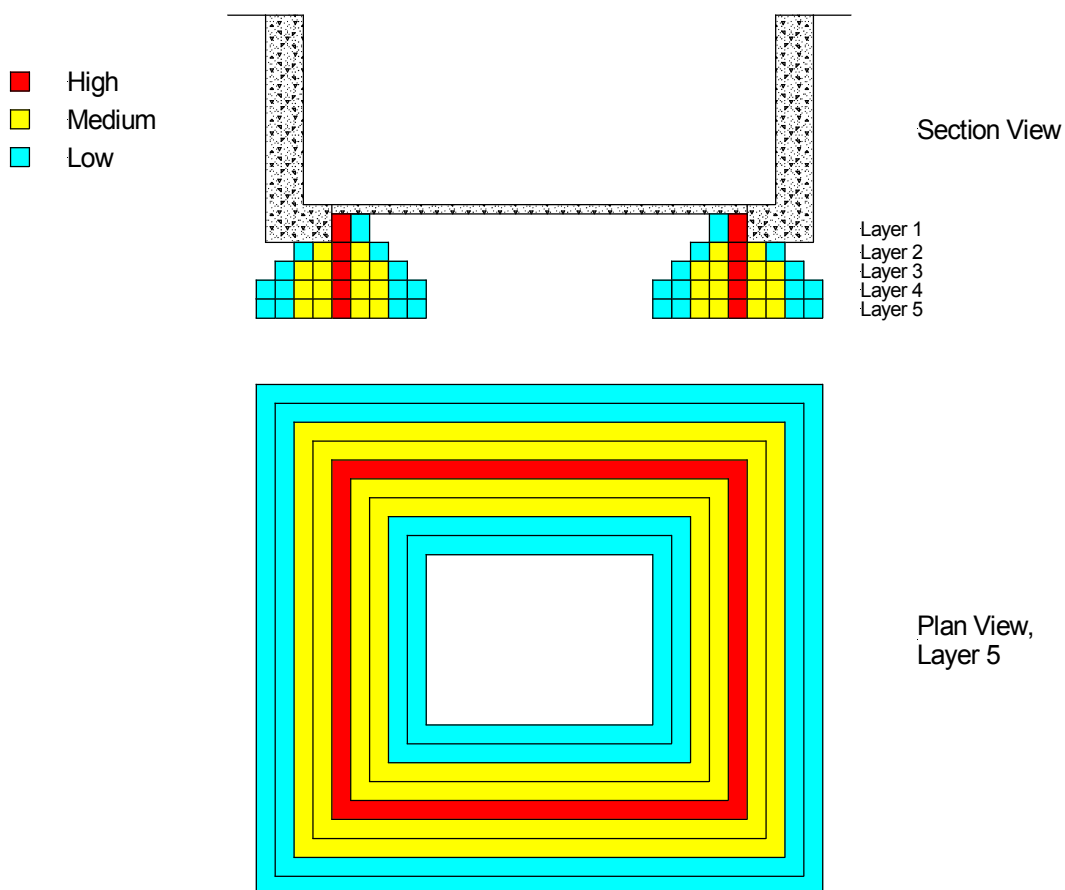

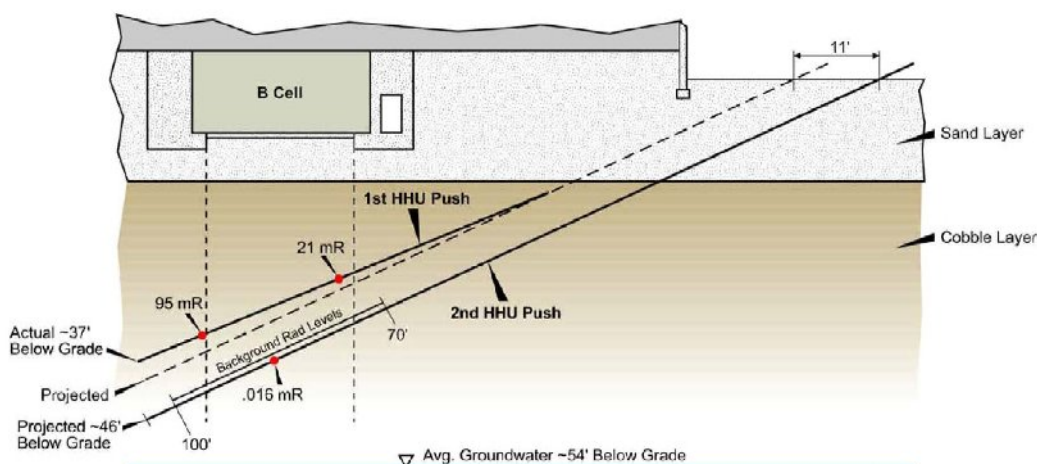**Figure 4. Simplified Contamination Field**

Table IV. Contamination Field Calculations

Zone	Volume (ft³)	Soil Contamination (Ci/ft³)	Cs-137 Activity (Ci)	Sr-90 Activity (Ci)	Total Activity (Ci)
High	429	3.69E+02	1.10E+05	4.84E+04	1.58E+05
Medium	1092	2.07E+00	1.57E+03	6.89E+02	2.26E+03
Low	1041	1.61E-03	1.16E+00	5.11E-01	1.68E+00
Total	2562		1.12E+05	4.91E+04	1.61E+05

HYDRAULIC HAMMER UNIT MEASUREMENTS

An additional campaign of exposure rate measurements was performed to determine whether the contamination had reached ground water. However, the method was modified to reach below the sand layer under B Cell and into a layer of cobbles that could not be penetrated by the original Geoprobe driver. Instead, a more powerful Hydraulic Hammer Unit (HHU) driver and associated heavier gauge instrument tubes were used to push through the sand layer at an angle and penetrate the cobbles.

As shown in Figure 5, the first instrument housing was pushed at a 25 degree angle to a depth of approximately 37 feet. The cobbles caused the instrument tubes to bend slightly at the interface. A peak exposure rate of 95 mR/hr was measured a depth of 33 feet below grade. A second instrument housing approximately 6 feet deeper than the first found no radiological contamination. The two readings verified that the contamination had not reached groundwater.

Figure 5. Hydraulic Hammer Unit Sampling Campaign

PHYSICAL SAMPLING

Physical samples of contaminated soil were obtained to verify the Geoprobe and contamination field activity calculations, the assumed composition of the radioactive material, and the variation of the contamination field with depth. Two specially designed tube-in-tube instrument housings with two-piece penetrators were driven into place. The inner penetrator was removed using the attached inner tube, and a sample collector was run down the outer tube to obtain soil samples. The sample collector was designed to limit the size of the sample to minimize the exposure received by the workers.

One of the samples (actually the second sample collected) was taken at Geoprobe elevation and as close to the B-Cell sump as possible. The other sample was taken four feet directly below the upper sample to help determine how the contamination plume changed with depth. The results from the physical sampling campaign are summarized in Table V.

Table V. Physical Sampling Results

Sample	ASR	Location	Cs-137 Activity (Ci/ft^3)	Sr-90 Activity (Ci/ft^3)
J1JD49	8888	Lower sample	2.33E-01	1.36E+01
J1JD50	8902	Upper sample, near B Cell sump	3.87E+02	1.83E+01

The physical samples verified the behavior predicted by ground water specialists that Cs-137 would be sequestered close to the B Cell floor, while the Sr-90 would be more mobile in the soil. The phsyical sample near the B Cell sump also agreed with the high zone activities in Table III.

ACTIVITIES BELOW THE FLOOR

Since the physical samples indicated that the Cs-137 is in fact sequestered close to the B Cell floor, the possibility was recognized that the Cs-137 concentrations several feet below the floor may be significantly less than those directly under the floor. That region could not be explored by direct-push instrument housings due to the building foundations. However, since the floor is only 6 inches thick, exposure rate measurements taken inside the cell could conceivably be used to determine the Cs-137 concentrations under the floor.

After extensive cleanout activities, the redisual activity within the cell was calculated in 2010 using a series of exposure rate measurements within the cell and associated MCNP models [12]. At that time, the magnitude of the soil contamination field was thought to be negligible compared to the activity remaining in the cell, and was ignored.

The MCNP models in Reference [12] were revised to include the possibility of significantl soil contamination, and the photon spectrum developed for the Geoprobe activity calculations was added to in-cell photon spectrum to model two different streams of radioactive material. The conjugate gradient solver used in the Geoprobe activity calculations was also used in place of the least-squares solver in Reference [12]. The methodology described in Reference [12], with these modifications, was re-performed to calculate both the residual activity remaining in the cell and the soil contamination immediately beneath the floor slab. The results are shown in Table VI.

Table VI. Source Activities

Source	Activity (Ci)	Reference [12] (Ci)
Soil Total	4.43E+05	
Cell Total	1.28E+04	2.20E+04
Grand Total	4.55E+05	2.20E+04

The revised modeling indicates that the activity remaining in the cell is approximately one half of the original estimate, and that there are 460 kCi of activity in the soil directly under the B Cell floor. This is in addition to the 160 kCi of activity at the level of the Geoprobes. The geographic distribtion of the activity directly under the B Cell floor is shown in Figure 6.

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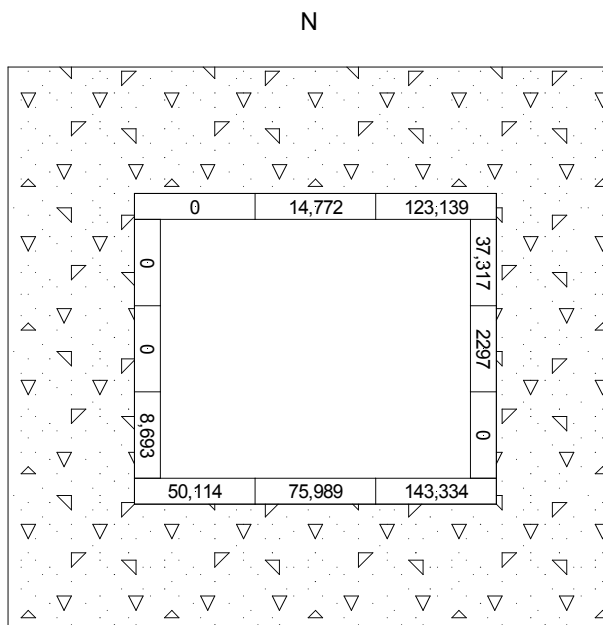
Figure 6. Activity Distribution

It is important to note that the revised modeling has a limited capability of distinguishing between relatively small activities in the cell and relatively large activities in the soil beneath the cell floor. The model is only able to make this distinction by utilizing the assumption that the activity on the floor of the cell is uniform, while that in the soil is non-uniform and concentrated in the vicinity of the expansion joint. Consequently, the calculated activities below the floor are much more uncertain that those associated with the Geoprobe measurements.

CONCLUSIONS

Based on the results of the sampling campaign, it is likely that the radiaoctive material below B Cell is primarily consists of feed solutions from the FRG Canister Fabrication Project, and that it contains purified Sr-90 and Cs-137 with enough actinide carryover to make some of the soil transuranic. The close agreement between the Geoprobe calculations and the physical samples adds confidence that there are more than 3700 TBq (100,000 Ci) of Sr-90 and Cs-137 in the soil approximately 1 meter below the cell floor. The majority of the Cs-137 is contained in the first meter of soil, while significant Sr-90 contamination extends to 10 meters below the cell floor.

It is also likely that an additional 15,000 TBq (400,000 Ci) of Cs-137 and Sr-90 activity is present directly below the floor of the cell, and that the residual activity inside the cell is only half of the previous estimates. However, the partitioning of activity between residuals in the cell and in the soil below the floor is much more uncertain than the activity calculations associated with the Geoprobe measurements.

Taken together, the calculated soil activities represent about half of the spill associated with the FRG Canister Fabrication project. The remainder of the spill is believed to have remained in the cell, where the majoriy has been removed as part of cell cleanup activities.

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The magnitude of the soil contamination below 324 B Cell is sobering, and it represents one of the most challenging remediation activities in the DOE complex. Of course, safe remediation begins with a good understanding of the magnitude of the problem. As a result, additional modeling and cross-comparison efforts are planned for 2012.

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