Evaluation of a Technetium-99 Detector Based on Laboratory Testing for Use in In-Situ Vadose Zone Applications

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Abstract: This document evaluates the feasibility of in-situ detection of technetium-99 in Hanford Site vadose zone soils (the soils between the surface and groundwater) using laboratory tests.

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Executive Summary

This document evaluates the feasibility of in-situ detection of technetium-99 (^{99}Tc) in Hanford Site vadose zone soils (the soils between the surface and groundwater) using laboratory tests.

The detector system performs adequately for high technetium concentration, but more development and laboratory testing is needed before field demonstration is performed.

The detector system clearly detects a 100.8 nanoCurie surface-deposited source of ^{99}Tc . The display system monitoring the tests shows that the detection system and the associated electronics can detect ^{99}Tc in the presence of potassium-40 (^{40}K) when the ^{99}Tc amount is large.

This report recommends that future work consist of

- Further testing using the current proof-of-principle detector system
- Discussions with Hanford Site staff on needed requirements for field testing and deployment
- Development and testing of a second (and possibly third) prototype detector
- Development of a new ^{99}Tc Detector System suitable for field testing
- Hanford Site laboratory and field testing of an integrated, ruggedized, field-ready, down-hole detector system ready for Hanford Site deployment

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1 Introduction

1.1 Purpose

The purpose of this document is to present the results of laboratory conducted proof-of-concept tests and to evaluate the feasibility of in-situ detection of technetium-99 (^{99}Tc) in Hanford Site vadose zone soils (the soils between the surface and groundwater) using laboratory tests.

1.2 Statement of Problem

Large (capacity of ~1,000,000 gallons) underground tanks at the Hanford Site have been used and are being used to store radioactive and chemically hazardous waste from the processing of irradiated nuclear fuel. Some of these tanks have released contamination into the soil.

Washington River Protection Solutions, LLC (WRPS) is under contract to the U.S. Department of Energy (DOE) to manage these tanks.

Because of its abundance, half-life, mobility, and toxicity, ^{99}Tc is considered a major contaminant of concern. However, ^{99}Tc is difficult to detect in-situ because of its low-energy radiation. Measurements of ^{99}Tc soil concentrations are usually obtained by collecting soil samples and measuring their concentrations in a laboratory.

This effort evaluates whether an in-situ ^{99}Tc detector can be developed. The first step (covered in this plan) is to perform laboratory measurements to determine if the radiation from ^{99}Tc decay can be detected and discriminated from the other radiation (mostly natural) contained in Hanford soils. If the first step is successful, an integrated ruggedized field-ready down-hole detector system will be demonstrated in the field as a direct push probe hole is decommissioned.

If the field demonstration is successful, the integrated ^{99}Tc detector system will join the set of tools used to characterize the vadose zone. After a direct push probe hole is pushed to the desired depth, gamma and neutron logging tools are used to determine the distribution of strongly gamma-emitting radionuclides and of moisture content, respectively. As the probe hole is decommissioned, the ^{99}Tc detector would be used to determine the distribution of ^{99}Tc in the vadose zone. With this information, depths can be selected at which samples would be obtained for laboratory analysis using a second direct push probe hole.

1.3 Background

Background information on the Hanford Site, past vadose zone characterization effort and methods, vadose zone concentrations, and the nuclear physics associated with the detector are documented in Section 1.3 of the *Evaluation Plan for an In-Situ Vadose Zone Technetium-99 Detector* (RPP-ENV-42008).

1.4 System Description

The system tested consists of a multi-element detector specific for ^{99}Tc , which is optimized to reduce background effects in the outer ^{99}Tc detector element, associated electronics (including a pulse shape discrimination system), and a power supply. The ^{99}Tc detector is composed of two

cylindrically coupled scintillators with different decay time constants. By discriminating on the pulse shape, one can distinguish interactions in either scintillator with a common photomultiplier tube. The system tested was not ruggedized nor configured for down-hole deployment.

However, the detector was sized to accommodate a 1.25-inch inside diameter push tube to ensure that detector efficiency and laboratory test results are consistent with requirements anticipated for field deployment.

1.5 Structure of the Document

This section provides background material for the evaluation of the ^{99}Tc detector. Section 2 presents the summary of the *Evaluation Plan for an In-Situ Vadose Zone Technetium-99 Detector* (RPP-ENV-42008). Section 3 gives the results of the laboratory testing. Section 4 provides an analysis of the laboratory testing and recommendations of future work. Section 5 describes the recommended path forward. Section 6 supplies the references used in this document. Copies of the laboratory notebook pages covering the preparation of samples and testing of the detector are maintained in the WRPS Quality Assurance records.

2 Summary of the Plan for Evaluation

The *Evaluation Plan for an In-Situ Vadose Zone Technetium-99 Detector* (RPP-ENV-42008) presents the requirements for the laboratory testing of the ^{99}Tc detector. It is summarized in this section.

2.1 General Approach

The general approach to evaluate the technetium-99 (^{99}Tc) detector is to determine how readings from the detector, taken in a laboratory setting, compare with calibrated point sources and with laboratory analyses of Tank Farm vadose zone samples. Samples were chosen so that the ^{99}Tc concentrations cover a wide range (~1 to 1,000 pCi of ^{99}Tc per gram of soil). The distance from the sample face to the detector face will be varied to determine the efficiency of the detector for differences in air distance between the soil and the detector. Finally, different soils will be used to determine the effect of non- ^{99}Tc radiation (such as naturally occurring potassium-40 – ^{40}K) on the results.

Note that for laboratory testing, the detector was positioned with its cylindrical axis horizontal to the laboratory bench surface. The technetium-containing soil was placed in a rectangular tray beneath the detector. Although such a geometry does not provide an accurate calibration for a in-situ environment, it does provide a test of how well the detector can distinguish between ^{99}Tc and natural background radiation, the effect of the air gap between the detector and the soil, and the effect of thin laboratory samples, whose thickness more closely mimic the effective depth for the vadose zone.

2.2 Sample Preparation

Following the requirements of RPP-ENV-42009, the Pacific Northwest National Laboratory (PNNL) prepared samples. With the concurrence of the WRPS, PNNL selected split-spoon liners previously obtained for WRPS as the source material for the soil samples. The characteristics of the samples are presented in Table 1.

The containers to hold the material are roughly rectangular trays having an inside length of ~5 inches, a width of ~3 inches, and an inside height of ~2.3 inches. The containers hold ~34.5 cubic inches (565 ml) and contain ~960 grams of soil.

The nominal 30, 100, and 500 pCi/g ^{99}Tc samples were shipped to the Idaho National Laboratory (INL) and used for the testing.

Table 1. Vadose Zone Sediments to Be Used to Create Testing Samples

	Desired Tc Concentration (pCi/g)					
	1	10	30 ^e	100	500 ^e	1000 ^e
Location	C-105	C-105	SX-115	T-106	T-106	T-106
Borehole Number	C4297	C4297	B8809	C4104	C4105	C4105
Sample ID	S04028-37A	S04028-86	B8809-25A	C4104-22B	S03044-12A/B	S03044-9A/B
Depth (ft bgs)	62.5	137	100	123	96.7	86.7
Tc-99 (pCi/g)	1.29	8.42	27.3	113	497	1050
K-40^e (pCi/g)	11.6	14	22.3	14.3	11.5	22.3
Co-60^e (pCi/g)	ND ^a	ND ^a	ND ^a	ND ^a	0.555	0.453
Cs-137^e (pCi/g)	ND ^a	ND ^a	ND ^a	ND ^a	0.039	0.028
Th-232^e (pCi/g)	ND ^a	ND ^a	ND ^a	ND ^a	ND ^a	ND ^a
U-238^e (pCi/g)	0.122 ^c	0.213 ^c	<1	0.13 ^c	6.58	<1.06
Nitrate (ug/g)	2.09 ^d	19.5 ^d	424 ^d	112 ^d	769 ^d	1030 ^d
Sodium (ug/g)	313 ^c	404 ^c	NM ^b	768 ^c	584 ^c	302 ^c
Actual Mass in Sample (g)	604	544	484	570	510	357

^a "ND" indicates the analyte was not detected in the sample.^b "NM" indicates the analyte was not measured in the sample.^c reported data was based on analysis of acid extract sample.^d reported data was based on analysis of water extract sample.^e indicates sample sent to INL for analysis.

2.3 Hardware Description

The hardware used in the testing is shown in Figure 1. The system components used for the testing are identified in Table 2. Photographs of the detector are shown in Figures 2, 3, and 4.

Table 2. MFC System Components

Manufacturer	Model No.	Description	Serial No.
Saint Gobain	1.512BC404 /3C444G/1.5-X	Detector	200-875 RJ776
ORTEC	556	High Voltage Power Supply	06219603
ORTEC	460	Delay line amplifier	2802
ORTEC	552	Pulse Shape Analysis/SCA	772-30
ORTEC	567	TAC/SCA	06221594
Fast Comtec	7072T	Dual ADC	60756
Fast Comtec	MPA-3	Dual Parameter MCA	331
DELL Computer	Precision 940	Computer	BYNN811
Microsoft	5.1	Windows	NA
Fast Comtec	MPANT	MPA-3 Analysis software	Version 1.67
Fast Comtec	Server	MPA-3 Server	Version 1.67

Figure 1. Hardware Diagram

The computer (a standard PC running MPANT Version 1.67 commercial software under the Microsoft 5.1 Windows Operating System) provided total counts in the inner and outer detectors. No cuts on energy or on coincidences were performed.

Figure 2. Picture of Probe (1 of 3)

Figure 3. Picture of Probe (2 of 3)

Figure 4. Picture of Probe (3 of 3)

3 Results of the Tests

INL received the detector system on August 14, 2009 from Saint Gobain Crystals (12345 Kinsman Road; Newbury, Ohio 44065) and performed acceptance testing, which was successful. From August 14 through August 24, 2009, INL staff performed functional performance testing of the system. Formal testing, witnessed by Fred Mann, Dave Myers, and Kathi Dunbar of WRPS was conducted on August 26 and 27, 2009. The laboratory notebook used was numbered 1347.

3.1 ⁹⁹Tc Point Source Testing

The testing was performed by Doug Akers (INL) on August 26 at Building IF-638 in Idaho Falls. It was witnessed by Kathi Dunbar, Fred Mann, and Dave Myers (WRPS) as well as by Denis Crass (North Wind Inc.). The detector system was placed on a laboratory benchtop. A 100.8 nanoCurie ⁹⁹Tc source was placed source down on top of the detector (in contact with detector) or on an adjustable laboratory stand (for other distances). The ⁹⁹Tc source is a surface mounted point source. Each counting period was approximately 200 seconds. Table 3 shows the counts for each measurement as the distance between the top of the ⁹⁹Tc source and the detector side was changed. All counts in the outer detector were attributed to ⁹⁹Tc. The distances between the source and the detector were measured with a rule, but because of the curvature of the detector and the sensitivity of the detector and source, the measurements are approximate. Figure 5 shows the relationship between distance and counts.

Table 3. Results of ⁹⁹Tc Point Source Measurements ^a

Distance from Source to Detector Face (inches)	Count Time (seconds)	Number of Counts Attributed to ⁹⁹ Tc
0	200.2	74,354
~0.3	200.1	42,269
~0.7	200.1	21,707
~1.4	200.0	13,410
~3	200.0	5,459
~6	200.0	2,419
No source	200.0	1,614

a Count duration was about 200 seconds. ⁹⁹Tc source strength was 100.8 nanoCuries.

Figure 5. Relationship between ^{99}Tc Point Counts and Distance from the Detector

3.2 ^{99}Tc Point Source and 30 pCi/g Soil Sample Testing.

Given the results of the previous test, the WRPS lead representative decided that this test was not needed. A test with Idaho soil was performed, showing clean separation between the ^{99}Tc peak and the ^{40}K peak. However, because Idaho soil derives from basalts, while Hanford soil derives from granite (which is higher in ^{40}K content), actually ^{40}K interference was evaluated using Hanford supplied soil samples (see Section 3.3).

3.3 Soil Sample Testing.

Testing was conducted by Doug Akers (INL) on August 27 at the Materials and Fuels Campus (MFC) of the Idaho National Laboratory. The laboratory room used has significant background. The testing was witnessed by Kathi Dunbar, Fred Mann, and Dave Myers (WRPS) as well as by Denis Crass (North Wind Inc.). The box containing the three soil samples (nominal 30 pCi/g ^{99}Tc , nominal 100 pCi/g ^{99}Tc , and nominal 500 pCi/g ^{99}Tc) shipped from PNNL was opened. As shown in Figure 6, the detector was placed on a pair of bismuth shielding blocks to support the detector on its horizontal axis. Each soil sample was placed on an adjustable labjack. Each counting period was about 500 seconds. Table 4 shows the counts for each measurement as the distance between the top of the nominal soil source and the detector side was changed. All counts in the outer detector were attributed to ^{99}Tc , while all counts in the inner detector were attributed to ^{40}K . The distances between the source and the detector were measured with a rule, but because of the curvature of the detector and the sensitivity of the detector and source, the measurements are approximate. Figure 7 shows the relationship between distance and counts.

At the end of this sequence of measurements, the detector stopped functioning. That is, no counts were detected.

Figure 6. Photograph of Soil Sample Counting

Figure 7a. Relationship between ^{99}Tc Counts from Soil Samples and Distance from the Detector

Figure 7b. Relationship between ^{40}K Counts from Soil Samples and Distance from the Detector **Table 4. Results of Soil Sample Measurements ^a**

Soil Sample	Distance from Soil Source to Detector Face (inches)	Count Duration (seconds)	Number of Counts Attributed to ^{99}Tc	Number of Counts Attributed to ^{40}K
500 pCi/g	~0.75	500.3	11,018	69,529
	~1.25	500.3	9,749	62,976
	~1.75	500.3	9,887	64,245
	~2.75	500.3	9,845	66,265
100 pCi/g	~0.75	500.3	10,233	66,226
	~1.25	500.3	10,041	67,700
	~1.75	500.3	9,835	65,007
30 pCi/g	~0.75	500.3	10,073	68,334
	~1.75	500.0	10,017	70,042

^a Count duration was about 500 seconds.

4 Analysis of the Testing and Recommendations on Future Work

4.1 Analysis of the Testing

4.1.1 ^{99}Tc Point Source Testing

Figure 5 displays the results of the first set of tests (point ^{99}Tc source as a function of space between the source and the detector face). As expected, the detector was most efficient with the point source on the detector. As the size of the air gap increased, the air deflected and slowed down the beta particles (electrons) emitted by the ^{99}Tc source. A distance of 0.3 inches reduced the efficiency by a factor of 2. The detector performed as expected.

4.1.2 ^{99}Tc Point Source Testing in the Presence of ^{40}K

The WRPS lead representative decided that this test as described in Section 3.1.2 was not needed.

4.1.3 Soil Sample Testing

The soil sample tests performed on August 27 produced unexpected results. Although the amount of ^{99}Tc in the soil samples was comparable to the 100 nCi ^{99}Tc point source (300 nCi for the 500 pCi/g soil sample, 60 nCi for the 100 pCi/g soil sample, and 18 nCi for the 30 pCi/g sample), the responses were quite different. Moreover, according to the PNNL analytical results, the ^{40}K concentration was a factor of 2 higher in the 30 pCi/g soil sample than in the 500 pCi/g sample, with the 100 pCi/g sample having a ^{40}K concentration in between. However, the results showed a consistent number of counts attributable to ^{40}K . Looking at the displays, there also seemed to be very high background, which was not reduced by shielding blocks.

The day after the WRPS-supervised testing, it was determined that the outer sheath of the detector was not anodized and therefore conductive. Supporting the detector on the bismuth blocks for the soil sample tests; i.e., contact between the bismuth block and detector, created a ground fault in the conductive (ungrounded) detector outer sheath (Figure 6). A photograph (Figure 8) shows a small crease on the detector surface where it contacted the bismuth block. It is believed that a ground loop had formed, causing the large background observed during the measurements as well as the failure of the detector to detect counts at the end of the August 27 soil tests.

Figure 8. Photograph of Detector Face after Soil Testing.

4.2 Additional Measurements

On August 28, 2009, Doug Akers repeated the ^{99}Tc point source measurements of August 26. The reproducibility was very good. Based on the assumption that the unsuspected ground loop was the problem, WRPS authorized a repeat of the soil sample measurements.

The soil sample measurements were repeated on September 1, 2009 at the MFC, but in a different room having lower background. This time the measurements also included the point ^{99}Tc source in the presence of the 30pCi/g soil sample. The electronics setup was the same as for the previous measurements. Figure 9 shows how the detector and soil sample was held to avoid ground loops. The count time was about 200 seconds. Table 5 shows the results of these experiments as well as the repeat of the soil measurements.

Time responses for the 30pCi/g and 500 pCi/g soil samples at 0.75 inch are shown in Figures 10 and 11. The time responses for the point ^{99}Tc source and the 30 pCi/g soil sample are shown in Figure 12, both nominally at 0.75 in from the detector.

Figure 9. Detector and Soil Samples Set-Up for September 1 measurements.

Table 5. Measurements Taken on September 1, 2009.
 Count Duration was 200 Seconds)

Spectrum ID	Description	Soil to detector distance (In.)	Outer Scintillator	Inner Scintillator
Bkgd_035	Background in vault	0.75	191	301
Tc-99-D500_036	500 pCi/g soil	0.75	510	582
Tc-99-D500_037	500 pCi/g soil	1.25	476	577
Tc-99-D500_038	500 pCi/g soil	1.75	447	505
Tc-99-D500_039	500 pCi/g soil	2.25	392	461
Tc-99-D500_040	500 pCi/g soil	2.75	376	480
INL Dirt_041	Normalized soil blank	0.75	242	404
Tc-99-D100_042	100 pCi/g soil	0.75	351	491
Tc-99-D100_043	100 pCi/g soil	1.25	312	471
Tc-99-D100_044	100 pCi/g soil	1.75	281	440
Tc-99-D100_045	100 pCi/g soil	2.25	300	411
Tc-99-D30_046	30 pCi/g soil	0.75	427	619
Tc-99-D30_047	30 pCi/g soil	1.25	322	510
Tc-99-D30_048	30 pCi/g soil	1.75	286	466
Tc-99-D30_049	30 pCi/g soil	2.25	251	446
Tc-99-D30_050	30 pCi/g soil w/o ⁹⁹ Tc source	0.75	374	514
Tc-99-D30_051	30 pCi/g soil w/ ⁹⁹ Tc source at 0.25 in	0.25	2323	575
Tc-99-D30_052	30 pCi/g soil w/ ⁹⁹ Tc source at 0.75 in	0.75	1236	594
Tc-99-D30_053	30 pCi/g soil w/ ⁹⁹ Tc source at 1.5 in	1.50	1072	625
Tc-99-D30_054	30 pCi/g soil w/ ⁹⁹ Tc source at 1.75 in	1.75	1578	544

Figure 10. Time Responses for the 30pCi/g Soil Sample

Figure 11. Time Responses for the 500pCi/g Soil Sample

Figure 12. Time Responses for the 30pCi/g Soil Sample and Point ⁹⁹Tc Sample

4.3 Observations of Additional Measurements

The following observations are central to the analysis:

- High background
- Repeatability
- Sensitivity
- Understanding the system

The room where the soil sample measurements were made had a high background (191 counts in 200 seconds in the outer detector and 301 counts in the inner detector) compared to 351 counts in the outer detector and 491 counts in the inner detector for the same time period for the 30 pCi/g soil sample 0.75 inches from the detector. Thus, a large fraction of the counts were simply from room background. Because of the short range of beta particles, all of this background is probably gamma radiation. **Thus, further laboratory testing should be performed in an environment having very low background.**

One measurement was repeated, 30 pCi/g soil at 0.75 inch from the detector (identification labels = Tc-99-D30_046 and Tc-99-D30_050). The ratios of the measurements are 1.17 ± 0.08 and 1.20 ± 0.07 for the outer and inner detectors respectively. The small departures from unity are most likely explained by having the soil sample at two slightly different distances from the

detector in the two cases. As shown in Figure 5, the efficiency of the detector is sensitive to this distance and for the purpose of these tests precise distance measurements were not required.

Thus, for future laboratory testing the distance should be fixed and not changed during soil sample replacement. Furthermore, a limitation (although not serious) of any in-situ detector system will be the amount of the air gap between the detector and the soil.

Approximately 10 percent of the detector area was exposed to the soil, the detector having been sized for actual field operation. The counts per pCi/g of soil in a 200 second measurement can be estimated by subtracting the counts from the 30 pCi/g and 100 pCi/g soil samples from the 500 pCi/g soil sample, resulting in 0.21 ± 0.08 (100 pCi/g base) to 0.34 ± 0.9 (30 pCi/g base) counts per pCi/g of soil in 200 seconds. These estimates are slightly low because some of the count can be ascribed to the higher ^{40}K in the 100 pCi/g soil sample (24% higher) and 10 pCi/g soil sample (94% higher). Thus, in a 200 second count, one would expect 2 to 3 counts for 30 pCi/g of soil for this configuration. Assuming the entire detector surface could be used, the total counts in a 200 second measurement would only be 20 to 30 counts. Also, it is expected that the distance between the soil and the detector will be closer than 0.75 inches, gain another factor of efficiency (perhaps a factor of 4). **Thus, the ability to see such levels is based on decreasing the amount of background caused by ^{40}K beta. Using the current detector system, the easiest way to perform this is to veto counts in the outer detector if a count in the inner detector is detected (say within 20 microseconds). Given the lower count rate observed for the 30 pCi/g soil at 0.75 inches from the detector (~5 per second), such coincident requirements are straight forward.**

Note that a much more accurate analysis of the sensitivity can be performed knowing the concentrations of the ^{40}K and ^{99}Tc in the soil samples. However, for this evaluation such an analysis is not warranted as it is clear from the testing performed that while the detector system has very large potential, further development is needed. The room background run shows that for every three counts in the inner detector, only 2 counts are seen in the inner detector. However, for a strong high energy beta source (e.g., the 500 pCi/g soil sample) the ratio of counts of the inner detector to the outer detector is 0.88 ± 0.12 (at 0.75 inches) and 0.97 ± 0.14 (at 1.25 inches). **However, for further development of this detector system, a good understanding of the detector system is crucial. Furthermore, understanding the degradation of the ^{99}Tc beta spectrum through the air gap and soil will be important in sizing the dimensions of the detector and setting the energy cuts.**

One further observation for field deployment should be noted. For laboratory tests, the count rate, as stated above, is quite low. However, there will be areas where the ^{137}Cs soil concentration will be quite high. Provisions should be made to eliminate the power to the photomultiplier tube whenever the count rates exceed some level (~1,000 counts per second) in order to save the photomultiplier tube.

4.4 Recommendations for Future Work

The detector system performs adequately for high technetium concentration. More development and laboratory testing of the current proof-of-concept system is needed before field demonstration is performed. A new detector should be designed and procured for a field demonstration. A laboratory demonstration at the Hanford Site should immediately precede the field demonstration so that DOE's Office of River Protection and Washington State Department of Ecology staffs can observe the detector performance in a controlled environment.

5 Recommended Path Forward

This section presents the future activities for the ^{99}Tc detector development and field testing:

- Further testing at INL using the current detector system (Section 5.1)
- Discussions with Hanford Site staff on needed requirements for field testing and deployment (Section 5.2)
- Development and testing of new ^{99}Tc prototype detector system(s) (Section 5.3)
- Development of a field deployable detector system (Section 5.4)
- Laboratory and field testing of an integrated ruggedized down-hole detector system at the Hanford Site (Section 5.5).

Section 5.6 presents a rough order magnitude (ROM) schedule and cost estimate for these activities. Note that these activities are not currently included in the near term baseline, and will be pursued only following formal DOE approval through a baseline change request process.

5.1 Further testing at INL using the current detector system

The electronics used in the current laboratory testing are unsophisticated. However, given the time between detector acceptance and WRPS-supervised testing, little effort was given to the software cut selection and the methods for ^{40}K and background subtractions. No effort was expended to use coincidence information in the analysis. Optimizing data collection and analysis methods should contribute to further reducing the minimum detection level for ^{99}Tc . Therefore, it is proposed to have PNNL ship the nominal 1, 10, and 1000 pCi/g ^{99}Tc soil samples to INL. INL would use these and the soil samples they already have (nominal 30, 100, and 500 pCi/g) to optimize the associated electronics and better determine the sensitivity of the detector.

The emphasis of the testing should be on improving the signal to noise ratio for ^{99}Tc and on understanding the response of the detector system to varying amounts of ^{99}Tc and ^{40}K .

Deliverable: At the end of this activity, the Detector Contractor will send to WRPS a letter report describing the testing and optimization of the detector system as well as lessons learned, including limitations of the system in the presence of non- ^{40}K beta sources.

5.2 Discussions with Hanford Site staff on needed requirements for field testing and deployment

The Hanford Site and tank farms in particular have stringent requirements for the use and release of instruments. Discussions should be held with

- Field work supervisors
- Industrial health
- Radiation Control
- Operators
- Electricians,

- Other Hanford Site Prime Contractors (particularly the Plateau Remediation Contractor and the Pacific Northwest National Laboratory),
- Other geophysical contractors, and
- Others.

The purpose of these discussions will be to establish the necessary requirements for

- 1) a fully functional and integrated down-hole detection system,
- 2) entry and release of instruments in tank farms and the 222-S laboratory, and
- 3) minimum requirements for conducting field and laboratory demonstrations at Hanford site facilities.

These requirements are to be established before the final design of the field deployable detector (Section 5.4).

Based on initial discussions, WRPS will develop straw man discussion points for a 2-day meeting of the interested parties. The Detector Contractor will participate in this meeting.

Deliverable: WRPS will issue a formal report on the requirements for a field deployable in-situ vadose zone detector for technetium-99.

5.3 Development and testing of a new ⁹⁹Tc prototype detector system

Preliminary testing by INL showed that the initial prototype detector requires some design improvements. At a minimum these include:

- Light scattering inside the detector
- Coupling between the inner and outer detector.

Based on the INL preliminary measurements using the point ⁹⁹Tc source as a function of distance from the photomultiplier tube (PMT), a drop off in efficiency (about a factor of 2) occurs when the point source is near the PMT versus when the source is at the other end of the detector. Thus, efficiency may be improved by using a PMT at each end of the detector. This would complicate the design, data collection, and analysis. However, the improvement in efficiency may be worth the complexity.

Based on the INL preliminary measurements using the point ⁹⁹Tc source at different locations on the detector's surface, different efficiencies were noted. Since the detector is nominally symmetric, these differences are likely a reflection of different coupling between the inner and outer detectors. This is not surprising as the creation of this coupling was changed during the manufacture process. As this detector was a first of a kind, such difficulties were expected.

Thus, development of another prototype detector is suggested to optimize shape, length and thickness of the outer detector. Design should start before the measurements on the initial prototype are completed (as much of the testing using the initial is focused on the optimization of the electronics outside of the detector).

Again, an important emphasis is improving the signal to noise ratio for ^{99}Tc and on understanding the response of the detector system to varying amounts of ^{99}Tc and ^{40}K .

Deliverables: Before procurement of the new prototype detector, the Detector Contractor will supply to WRPS the goals and proposed test plan for the new prototype detector. At the end of activity, the Detector Contractor will send to WRPS a letter report describing the testing and optimization of the detector system as well as lessons learned.

5.4 Development of a field deployable detector system

Based on the information learned from further testing of the new system (Section 5.1), the requirements for deployment at the Hanford Site (Section 5.2), and the testing of a second prototype detector, a ruggedized new detector for Hanford Site field testing in a direct push probe hole would be developed. Work scope associated with this activity will also include development of all necessary ancillary hardware, analytical software, software validation, data management and control processes, and required field procedures to deploy and fully demonstrate an integrated ruggedized ^{99}Tc vadose zone detector system. Note that a fully integrated ruggedized ^{99}Tc vadose zone detector system includes

- 1) all detector components (e.g., detector scintillators, voltage divider, photomultiplier tube),
- 2) required analytical software and hardware,
- 3) all ancillary support equipment such as deployment mechanisms, umbilical management systems, and
- 4) necessary QA/QC to verify and validate analytical software and results.

Deliverable: The Detector Contractor will transport to the Hanford Site a detector system ready for laboratory and in-situ vadose zone field testing. The detector system will meet all requirements identified in the requirements document created above (Section 5.2). The Detector Contractor will send a WRPS a letter report describing the design, testing, and optimization of the detector system as well as lessons learned.

5.5 Laboratory and field testing of a field deployable detector system at the Hanford Site

Using the 3 remaining soil samples being stored at PNNL (nominally 30, 100, and 500 pCi/g of ^{99}Tc), the soil sampling test would be repeated using the field deployable detector. Such testing would allow staff for DOE's Office of River Protection and from the Washington Department of Ecology to observe testing of the vadose zone deployable detector in a controlled setting. Such testing would also provide information on count times needed in an in-situ vadose zone environment. Given the background and space issue identified at the 222-S Laboratories, this demonstration laboratory test should be performed at PNNL. Note that during the discussions with Hanford site staff requirements will be developed for testing and demonstration of equipment in Hanford laboratory facilities.

Field testing is anticipated to be accomplished through currently available direct push casing. Two elements are needed to optimize the test:

1. A range of ^{99}Tc activities over a reasonable depth in the vadose zone; and
2. Ease of access to the test site, by all parties.

Tank farms, due to the numerous restrictions on access, does not provide the optimal site for the initial field demonstration of the ^{99}Tc detector. Although the tank farms provide an environment where a wide range of ^{99}Tc activity is present throughout the vadose zone, the challenges of gaining farm access by non-Tank Farm personnel may be sufficient to remove them as preferred initial sites.

Outside the tank farm environment, one Hanford location, the BC Cribs and Trenches Site, provides a range of known ^{99}Tc activity dispersed vertically within the vadose zone. Technetium-99 activity ranges from less than detectable to as high as 145 pCi/g in borehole C5923 (Serne et. al., 2009). This borehole is associated with a deep vadose zone remediation experiment being conducted by the Plateau Remediation Contractor (PRC). The borehole exhibits two zones of elevated ^{99}Tc , an upper zone ranging from ~28 ft below ground surface (bgs) to ~166 ft bgs, and a lower zone ranging from ~210 ft bgs to 246 ft bgs. The upper zone is the subject of the remediation experiment/demonstration. Table 6, from Serne et.al., provides a distribution of ^{99}Tc in this region. A position adjacent to borehole C5923 or another equivalent borehole nearby (such as C5924 and C7047) is proposed for testing of the prototype down-hole ^{99}Tc detector. There is no intent to interfere with the on-going deep vadose desiccation experiment that uses Borehole C5923.

The test design of the down-hole demonstration of the ^{99}Tc detector will be such that it parallels how such a system will be typically deployed in a tank farm environment. An initial direct push will be made to a 115 foot depth. This hole will be logged for moisture content distribution and gamma emitters using a bismuth-germanium oxide probe. As this borehole is decommissioned, the prototype ^{99}Tc detector will be deployed at nominally 1 ft intervals. Following analysis of the moisture, gamma and ^{99}Tc logs, a decision will be made on whether or not to proceed with sampling of selected vadose zone horizons. This decision will be made jointly with representatives of the deep vadose zone remediation project to ascertain where samples should be collected.

Should sampling be deemed appropriate: sample depths will be determined and a second direct push initiated to collect samples from the selected depths. Analysis of the samples would then be conducted.

Deliverable: WRPS will issue a report describing the results of the laboratory and field testing along with a recommendation on whether or not to deploy the detector as part of the Tank Farm vadose zone characterization activities.

Table 6. Distribution of ⁹⁹Tc in Borehole C 5923

HEIS #	mid depth	Technetium-99	HEIS #	mid depth	Technetium-99
	ft bgs	pCi/g		ft bgs	pCi/g
BIT740	5.5	<0.424	BIT793	173	<0.424
BIT741	8	<0.424	BIT9L6	176.8	<0.424
BIT743	13	<0.424	BIT9L7	179.3	<0.424
BIT744	15.5	<0.424	BIT9L8	181.8	<0.424
BIT745	18	<0.424	BIT9L9	184.3	<0.424
BIT746	20.5	<0.424	BIT798	195.5	0.43
BIT816	20.5	<0.424	BIT7B4	210.5	85.09
BIT747	23	<0.424	BIT7B6	215.5	130.87
BIT748	25.5	<0.424	BIT7C2	230.5	82.53
BIT749	28	7.23	BIT9K6	236.8	71.19
BIT750	30.5	4.82	BIT9K7	239.3	50.81
BIT751	33	0.59	BIT9K8	241.8	38.88
BIT825	36.8	72.14	BIT824	244.3	24.16
BIT826	39.3	14.37	BIT7C4	245.5	14.71
BIT828	44.3	22.65	BIT7C9	258	0.55
BIT752	45.5	145.85	BIT7D1	263	<0.424
BIT753	48	93.57	BIT7D8	280.5	<0.424
BIT755	53	33.80	BIT7F2	290.5	<0.424
BIT757	58	46.71	BIT7H1	313	<0.424
BIT763	73	71.66	BIT7J2	340.5	<0.424
BIT766	80.5	93.25	BJV530	344	<0.388
BIT767	83	54.08	BJV531	346.5	<0.388
BIT817	83	43.86	BJV532	349	<0.388
BIT985	89.3	60.15	BJV533	351.5	<0.388
BIT768	90.5	74.75			
BIT771	98	34.30			
BIT773	103	28.10			
BIT9K9	106.8	26.88			
BIT9L0	109.3	44.54			
BIT9L1	114.3	20.13			
BIT777	123	63.31			
BIT818	123	56.88			
BIT9L2	126.8	66.03			
BIT9L3	129.3	38.65			
BIT9L4	131.8	44.29			
BIT9L5	134.3	89.99			
BIT781	143	51.68			
BIT790	165.5	0.44			

(a) <values = below level of quantitation.

(b) Red type = values above natural background and bold red are peak values in various lobes of vertical distribution.

5.6 ROM Schedule and Cost for Recommended Path Forward

The schedule and rough order of magnitude (ROM) cost of the recommended path forward (Sections 5.1 through 5.5) is displayed in Table 7. It should be noted that the recommended path forward has been designed to provide a progressive and systematic approach to completing development of the vadose zone ^{99}Tc detector system and ultimately a full scale demonstration of a field ready system at Hanford. At each step of the development process (sections 5.1 and 5.3 – 5.5) the Detector Contractor is required to present results and demonstrate successful achievement of objectives. At each of these gates WRPS will evaluate the results, provide comment as to recommended path forward, and authorize progression to the next step. Note that the schedule shown indicates duration and sequence, if this project were to start October 1, 2009. If this work is authorized through DOE approval of a baseline change request, the actual schedule will be finalized to reflect the work authorization date.

Table 7. ROM Schedule and Cost for Proposed Path Forward

Activity	Schedule		ROM Cost	
	Start	End	Organization	Cost (K\$)
5.1 Further testing at INL using the current detector system	Oct 09	Dec 09	Detector Contractor	110
			WRPS ^a	10
5.2 Discussions with Hanford Site staff on needed requirements for field testing and deployment ^b	Dec 09	Jan 10	WRPS ^c	30
			Other Hanford ^d	0
			Detector Contractor	5
5.3 Development and testing of a new ^{99}Tc prototype detector system(s)	Dec 09	Apr 09	Detector Contractor	350
			WRPS ^a	10
5.4 Development of a vadose zone deployable ^{99}Tc detector system	Mar 10	Aug 10	Detector Contractor ¹	460
			WRPS ^a	25
5.5a Laboratory and field testing of a new detector system, ready for Hanford Site deployment	Sep 10	Nov 10	Detector Contractor	24
			Laboratory Test ^e	20
			Other Hanford ^d	0
			Direct Push Support ^f	80
			CHPRC ^g	50
5.5b Analytic analysis of soil samples to verify field measurements	Nov 10	Feb 11	WRPS ^a	30
			Analytic Analysis ^h	20

ROM = rough order of magnitude

Assumptions:

- a) Mainly project management costs
- b) The requirements meeting will be 2 days in duration

- c) WRPS costs includes salaries for all WRPS staff at the requirements meeting plus preparation and document costs.
- d) Other Hanford (including contractors, DOE, and regulators) will participate at their expense as part of integration.
- e) Costs for the laboratory test assume the use of a laboratory for 1 week (2 day preparation, 2 day demonstration, and 1 day further test and take down).
- f) Includes all usual WRPS costs associated with a 4 week direct push campaign (1 day mobilization, 1 week for the initial push as well as gamma and moisture logging, 1 weeks for decommissioning including 1 foot technetium probe logging, 1 week for obtaining samples, and 2 days demobilization).
- g) CHPRC costs as field work will be on Hanford land managed by them.
- h) Costs for analysis of 3 samples taken from field test.
- i) Final cost and schedule for laboratory testing and field demonstration may be impacted based on functions and requirements established in b.

6 References.

- RPP-ENV-42008, F.M. Mann, K.J. Dunbar, C.F. Brown, Dennis Crass, and Douglas Akers, *Evaluation Plan for an In-Situ Vadose Zone Technetium-99 Detector*, RPP-ENV-42008, Washington River Protection Solutions, LLC, Richland, Washington.
- Serne et al., 2009. R.J. Serne, D.F. Rucker, A.L. Ward, D.C. Lanigan, W. Um, M.W. Benecke, B.N. Bjornstad, *Electrical Resistivity Correlation to Vadose Zone Sediment and Pore-Water Composition for the BC Cribs and Trenches Area*, PNNL-17821, Pacific Northwest National Laboratory, Richland, Washington.