Determination of the Distribution and Inventory of Radionuclides within a Savannah River Site Waterway - 13202

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ABSTRACT

An investigation was conducted to evaluate the radionuclide inventory within the Lower Three Runs (LTR) Integrator Operable Unit (IOU) at the U.S. Department of Energy's (DOE's) Savannah River Site (SRS). The scope of this effort included the analysis of previously existing sampling and analysis data as well as additional streambed and floodplain sampling and analysis data acquired to delineate horizontal and vertical distributions of the radionuclide as part of the ongoing SRS environmental restoration program, and specifically for the LTR IOU program. While cesium-137 (Cs-137) is the most significant and abundant radionuclide associated with the LTR IOU it is not the only radionuclide, hence the scope included evaluating all radionuclides present and includes an evaluation of inventory uncertainty for use in sensitivity and uncertainty analyses. The scope involved evaluation of the radionuclide inventory in the P-Reactor and R-Reactor cooling water effluent canal systems, PAR Pond (including Pond C) and the floodplain and stream sediment sections of LTR between the PAR Pond Dam and the Savannah River.

The approach taken was to examine all of the available Sediment and Sediment/Soil analysis data available along the P- and R-Reactor cooling water re-circulation canal system, the ponds situated along those canal reaches and along the length of LTR below Par Pond dam. By breaking the IOU into a series of sub-components and sub-sections, the mass of contaminated material was estimated and a representative central concentration of each radionuclide was computed for each compartment. The radionuclide inventory associated with each sub-compartment was then aggregated to determine the total radionuclide inventory that represented the full LTR IOU.

Of special interest was the inventory of Cs-137 due to its role in contributing to the potential dose to an offsite member of the public. The overall LTR IOU inventory of Cs-137 was determined to be 2.87E+02 GBq, which is similar to two earlier estimates. This investigation provides an independent, ground-up estimate of Cs-137 inventory in LTR IOU utilizing the most recent field data.

INTRODUCTION

In 2010 a Composite Analysis (CA) of the DOE's SRS was completed in accordance with DOE Order 435.1, Radioactive Waste Management. As required in the order, the CA evaluated the dose impact of the anticipated SRS end state residual sources of radionuclides to offsite members of the public. The CA evaluated doses to members of the public at the locations where SRS site streams discharge into the Savannah River. Although the models developed to perform this computation indicated that the dose constraint of (300 uSv/yr) associated with the CA was not approached at any of the points of assessment (POAs), the model indicated that the highest maximum dose was realized at the POA located at the mouth of Lower Three Runs Creek. The LTR creek and floodplain is one of six SRS Integrator Operable Units (IOUs) corresponding to

each of the major onsite watersheds. IOU's are defined as surface water bodies and associated wetlands, including the sediment, floodplain soil, and related biota. IOUs are evaluated to determine if past releases of contamination from SRS pose unacceptable risks to the environment, SRS workers or off-site members of the public. Radionuclide releases associated with IOUs are adsorbed to the canal, pond and streambed sediments and the soils of the shallow floodplains that lie immediately adjacent to stream channels. In accordance with the DOE's Low Level Waste Federal Facility Review Group (LFRG) review, which provided conditional approval to the SRS CA in 2010 on the condition that eighteen observations would be addressed, this work was conducted. The work was conducted to improve the CA sensitivity and uncertainty analysis in particular by improving the CA radionuclide inventory estimate. This report documents an investigation that was conducted to update the existing radionuclide inventory estimate for the soil and sediment associated with the LTR IOU at the SRS. [2] [3].

The scope of this effort included the analysis of previously existing sampling and analysis data as well as additional streambed and floodplain sampling and analysis data acquired to delineate horizontal and vertical distributions of the radionuclide as part of the ongoing SRS environmental remediation program, and specifically for the LTR IOU program. While Cs-137 is the most significant and abundant radionuclide associated with the LTR IOU it is not the only radionuclide, hence the scope included evaluating all radionuclides present and includes an evaluation of inventory uncertainty for use in sensitivity and uncertainty analyses. The scope involved evaluation of the radionuclide inventory in the P-Reactor and R-Reactor cooling water effluent canal systems, PAR Pond (including Pond C) and the floodplain and stream sediment sections of LTR between the PAR Pond Dam and the Savannah River.

DESCRIPTION OF LOWER THREE RUNS INTEGRATOR OPERABLE UNIT

The LTR IOU is located in the southeastern portion of SRS. P-Area Reactor and R-Area Reactor are the two primary SRS facilities located within the LTR watershed. The location with respect to the SRS boundaries is illustrated in Figure 1. In addition, the watershed contains a 2,500-acre lake (PAR Pond) used by both P- and R-Reactors during their active operation periods. From the PAR Pond Dam, LTR flows approximately 17 miles south to southwest before it enters the Savannah River at the boundary of SRS. This section of LTR contains a long, narrow strip of land known as the LTR tail, which is also owned by the DOE.

METHOD

The first step in the investigation was to obtain all of the available sample data from the LTR IOU by sub-compartment (i.e. P-Reactor cooling water effluent canal system, R-Reactor cooling water canal system, PAR Pond (including Pond C), and from the LTR reaches below the PAR Pond dam (see Figures 2 and 3). As part of their IOU program, SRS has maintained a database of Sediment and Sediment/Soil (Sed/Soil) samples that have been collected in support of their IOU program for SRS. These are the two types of samples that have been collected of contaminated media in the LTR IOU that were evaluated in this investigation. "Sediment" samples are defined as samples of soil collected from below a water surface body, whether that occurs in a pond, lake or stream. "Sed/Soil" samples are defined as samples that are not submerged beneath water, but are usually collected directly adjacent to ponds, lakes and streams.

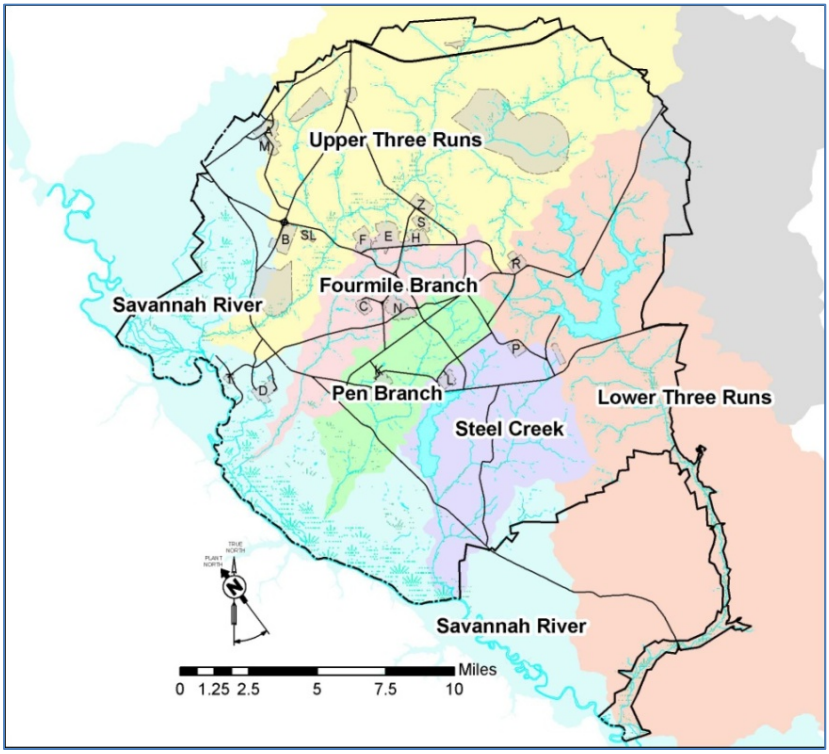

Figure 1 Location of Lower Three Runs Watershed at SRS

The set of samples associated with the LTR IOU were downloaded from the IOU project server using the Geographic Information System (GIS) program ArcMap. Use of ArcMap software is useful in that sample locations can be posted and examined within a geo-referenced view. Once the downloaded data was displayed within ArcMap, Sediment and Sed/Soil samples could be selected by LTR IOU sub-compartment, exported from ArcMap and read into Excel spreadsheets for analysis. Descriptions of the sampling programs conducted to acquire all of the data in support of the LTR IOU can be found in the four reports issued to describe different phases of the LTR IOU sampling program. These include [8] [9] [5] [6].

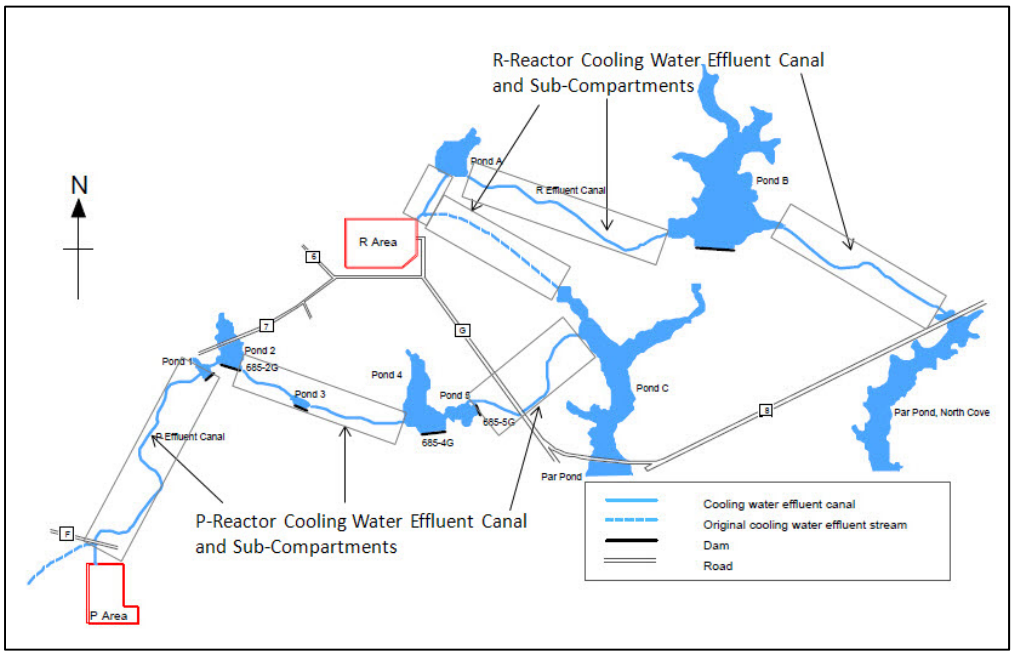

Figure 2 **P- and R-Reactor Canal System, Ponds and Sub-Compartments**

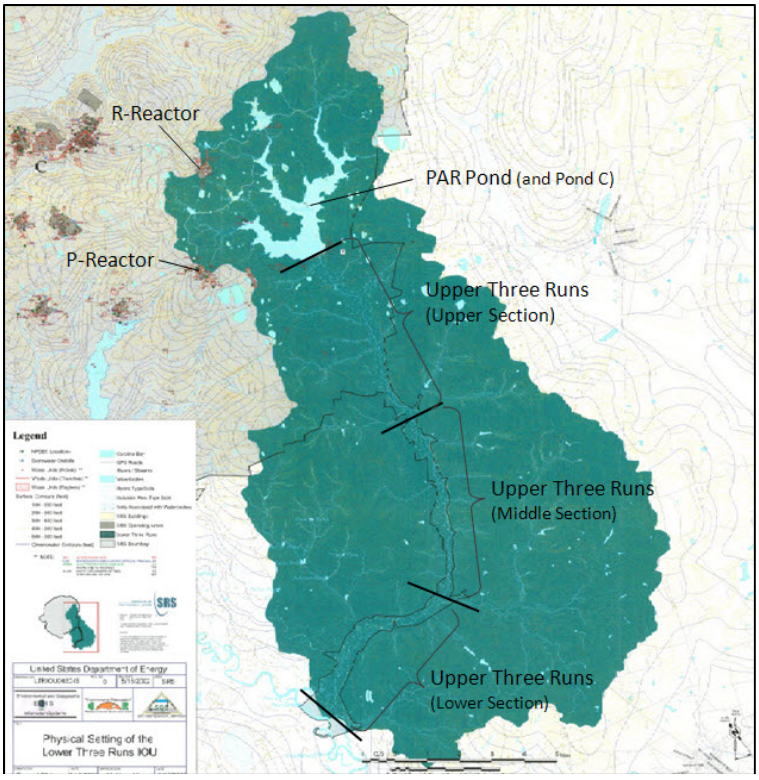

Figure 3 **Delineation of LTR Sub-Compartments below Par Pond Dam**

The ability to define separate sub-components of the LTR IOU (i.e. P-Reactor cooling water effluent canal system, R-Reactor cooling water effluent canal system, PAR Pond (including Pond C), and the LTR reaches below PAR Pond dam to the Savannah River) and to further sub-divide each sub-compartment into smaller sub-sections, as needed, was critical for developing a radionuclide inventory for the entire LTR IOU. The basic approach was to:

- Identify separate sub-components of the IOU.
- Identify separate sub-sections of each sub-component.
- Select and export the Sediment and Sed/Soil data for each sub-section.
- Categorize data into spreadsheets to evaluate individual radionuclides for each sub-section.
- Calculate representative concentrations for all radionuclides associated with the Sediment and Soil/Sediment samples in each sub-section.
- Estimate the volume and mass of contaminated soil for each sub-section.
- Compute the inventory of each radionuclide found to exist in each sub-section.
- Total the inventory from all sub-sections for each sub-component and then total the inventory from all sub-components for the total LTR IOU inventory.

Issues related to the use of sample analysis data

As previously mentioned, the Sediment and Sed/Soil analytical results were exported from ArcMap and read into Excel files for processing. Once in Excel, the data for a particular sub-compartment/sub-section was segregated by radionuclide within separate worksheets so that the estimate of a central, representative value for soil or soil/sediment activity concentration (Bq/kg) could be computed.

The radionuclides of interest, for which analyses were performed, include the following: Cs-137; Co-60; Sr-90; Cm-244; Cm-243/4; Am-241; Pu-239; Pu-238, Np-237; U-238; U-235; U-234; and U-233/4. In this investigation it is assumed that the laboratory analyses that indicate a result for Cm-243/244 and U-233/234 actually reflect activity levels for Cm-244 and U-234, respectively. It is assumed that if Cm-243 and U-233 contribute to the overall activity level in the analysis, that it is only a very minute proportion. Hence, in the radionuclide inventory rollup, activity levels from the “Cm-243/244” designation are rolled-up into the Cm-244 total and activity levels from the “U-233/234” designation are rolled-up into the U-234 total.

The U-233/4, U-234, U-235 and U-238 analyses in the LTR IOU database were evaluated to determine the central concentration values for each sub-compartment. These representative concentrations of each of the Uranium isotopes were then compared to the 2X average background values for Uranium isotopes from R-Area (41.9 Bq/g for U-233/234, 2.1 Bq/g for U-235 and 37.8 Bq/g for U-238) [7]. The natural uranium isotope curie ratio from R-Area was also used for comparison (0.495 for U-233, 0.023 U-235 and 0.483 for U-238) [7]. The results for individual compartments consistently showed that the ratios of the three different isotopes and the levels of activity reflect the natural occurrences of Uranium. Therefore, no inventories are reported for these isotopes because they represent natural levels.

Since the computation of radionuclide inventories depend heavily upon the laboratory analytical results associated with sediment and soil samples, it is important to understand the qualifiying conditions associated with each analytical result. The qualifiers in question include the following:

U – species/isotope was analyzed for, but not detected

J – reported value is an estimate

NQ – no qualification is associated with the result

Negative concentrations or activity levels are frequently reported in the analytical results and reflect the condition where the measured result is lower than the long-term background level that has been established for the specific instrument used to make the measurement and, in certain circumstances, are retained for computing representative central values (e.g. mean or median) of the radionuclide.

The rules and logic to selecting data analyses for inclusion in the calculation were obtained via verbal communication with subject matter experts who are employed in other ongoing environmental monitoring and reporting efforts at SRS, and who employ the same logic in those investigations. For a group of analysis results associated with samples collected in a particular sub-compartment (or sub-section) of the LTR IOU the following rules were applied:

- If the presence of radionuclide is confirmed in any analytical result, then all other sample results, including U-qualified samples and negative count results are included in the calculation of the representative central value. The rationale is that the U and (-) counts are still the best estimate for that radionuclide in that sample and cannot be censored by elimination from the calculation of the representative central value.
- If all sample results for a particular radionuclide in a particular sub-compartment/sub-section have U qualifiers (< detect), then the presence of that radionuclide has not been confirmed and it is presumed to not be present.
- A negative (-) results mean that the count for a particular sample was less than the background level determined over a long period of time for the analytical instrument.

Determination of the central, representative value of the analytical results for each radionuclide in a sub-compartment/sub-section in this investigation refers to the computation of the mean or median values. Virtually all of the computed mean values were higher than computed median values. Mean values were selected for use in computing the radionuclide inventories in order to avoid under-estimating the actual radionuclide inventory for a given sub-compartment/sub-section. There were a couple of instances in which the mean value for all analyses of a particular radionuclide was computed to be a small negative number. In those cases the median value was used as the representative central value, instead.

The Sediment and Sed/Soil samples utilized in this investigation were collected on different dates in the past. Many samples are much older than more recently acquired samples. This disparity was dealt with by standardizing the results by decay correcting all individual results to a reference date taken to be September, 2012. This process is particularly important to assess radionuclides with relatively short half-lives, like Cs-137. One exception to the standardization procedure is for Am-241. The presence of Am-241 in the LTR IOU is not the result of direct releases of that radonuclide from the P- and R-Reactors but rather from ingrowth from the decay of very small amounts of Pu-241 present in the Sediment and Sed/Soil. Thus, in this analysis it was not appropriate to decay Am-241 to September 2012 and the analytical results were used directly.

This results in a slight underestimation of Am-241 in LTR IOU, however Am-241 exists only in very low concentrations within LTR IOU to begin with, and therefore is not a risk driver.

Estimating the Mass of Contaminated Sediment and Sed/Soil

Once a representative activity level of each radionuclide was determined for contaminated Sediment and Sed/Soil associated with each of the LTR IOU sub-compartments/sub-sections, the mass of contaminated media associated with each sub-compartment/sub-section was estimated.

The parameters required to determine the mass of contaminated material included those to first estimate the volume of contaminated Soil or Sed/Soil in each sub-compartment/sub-section includes the following:

- Representative length
- Representative width
- Representative depth of the sample set
- Representative area and perimeter for ponds
- Representative bulk density of the soil/sediment material

The representative length and widths for Sediment and Sed/Soil zones within each sub-compartment of the canal systems or within the LTR waterway below the PAR Pond dam were estimated using the Measuring tool within ArcMap and careful examination of the Light Detection and Ranging (LIDAR) land surface contour coverage within the ArcMap project.

In addition to this, the availability of an above ground survey method employing LaBr detectors was employed in the LTR floodplain below Par Pond dam to estimate the width of contaminated swath of Sed/Soil on either side of the LTR waterway. Using this information, as well as the transect Sed/Soil analytical results, the representative combined widths of the two swaths of contaminated Sed/Soil on either side of LTR was established as 150 m.

The reported sample depth associated with Sediment (submerged) samples collected as part of the LTR IOU program is 0.2 m, which is the depth utilized both above and below Par Pond Dam, for canal, pond and LTR waterway submerged sediments. The use of this depth is justified by the depth-discrete samples collected within Pond B, which indicate that virtually all of the Cs-137 activity resides in the upper 20 cm. The basis for selecting the representative depths for use in estimating the mass of contaminated Sed/Soil was the recommendation provided by ACP. A depth of 0.35 m was utilized below Par Pond Dam while 0.3 m was utilized for Sed/Soil zones above Par Pond Dam. While historic sampling in the LTR IOU below the dam indicates that virtually all of the Cs-137 activity (and other radionuclides as well) resides in the upper foot of Sed/Soil zones, the effective depth of 0.35 m was utilized along these reaches in this investigation to account for the fact that some deeper samples (1-4 ft. interval) indicate a low level of activity. More detailed sampling information is available in the LTR Periodic Reports for the LTR IOU e.g. [5] [6].

Unique situation in Par Pond Sediments

In evaluating the analytical data associated with Par Pond, it became apparent that the Sediment could not be treated as a single zone with a single representative central concentration for the radionuclides. In particular, this was an issue with Cs-137. It was discovered that several distinct

hot spots of Cs-137 activity exist in the sediments beneath Par Pond. The location of these hot spots is indicated in Figure 4. These five locations contain the only Cs-137 activity levels in Par Pond sediments that are > 5.4 kBq/g, and which represent 3% of all Cs-137 analytical results for Par Pond. It was discovered that combining the highly elevated sample results from the hot spots with all other Cs-137 analytical results for Par Pond to compute the central, representative value for all Par Pond sediments results in a grossly over-estimated Cs-137 inventory. Since there is insufficient sample data to more accurately delineate the extent of the submerged hot spots and to treat them independently, a simpler approach was employed to obtain a more accurate Cs-137 inventory. This approach was to sub-divide Par Pond sediments into two zones, the Upper Par Pond reach, which contains all five hot spots, and the Lower Par Pond reach. Each of these zones was treated independently. Sample results were grouped separately for each of these zones for the purpose of computing the central, representative sediment concentration for each radionuclide, including Cs-137. The volume and mass of contaminated sediment associated with the Upper and Lower Par Pond reaches was computed using a factor of 0.5 of the total computed Par Pond sediment volume and mass. The total radionuclide inventories from both zones were then added together for the Par Pond total radionuclide inventory. In light of the impracticality of conducting an extensive sampling program to better define the extent of the submerged Cs-137 hot spots, this approach is considered adequate for estimating the Par Pond Cs-137 inventory.

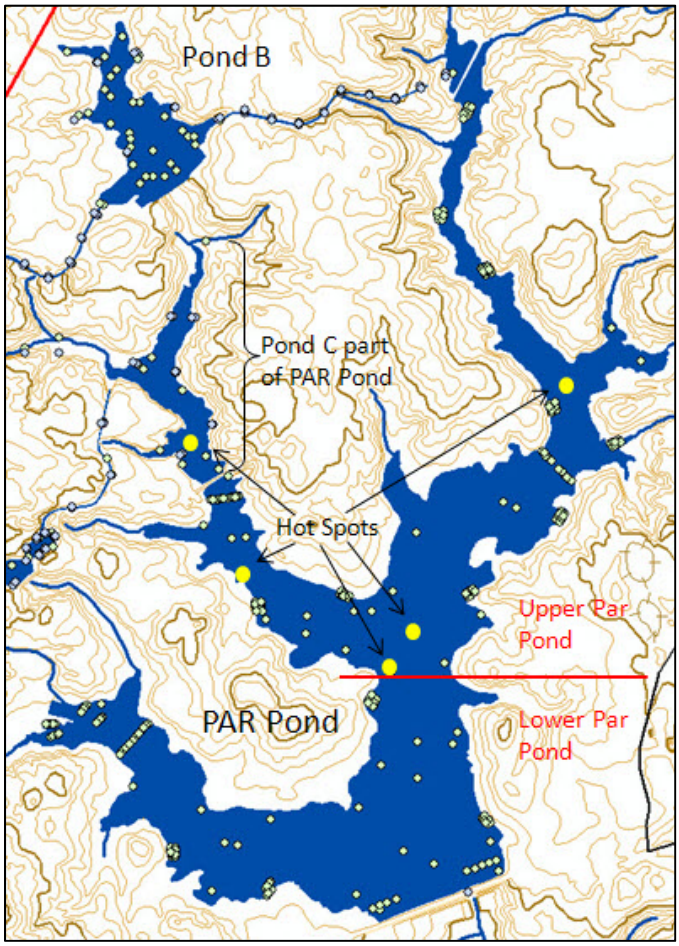

Figure 4 Upper and Lower Par Pond Segments and location of hot spots

RESULTS

The results reported for radionuclide inventories within LTR IOU were obtained for each of the main sub-compartments of LTR IOU. The values were obtained by aggregating the inventories calculated for each of the sub-sections associated with each sub-compartment. The total radionuclide activities are presented in Table I.

Table I. Radionuclide Inventory Roll-up for LTR IOU and its Sub-Compartments

	R-Canal System (GBq)	P-Canal System (GBq)	PAR Pond (GBq)	LTR below PAR (GBq)	Total for LTR IOU (GBq)
Cs-137	5.33E+02	7.03E+00	1.29E+03	8.07E+02	2.63E+03
Co-60	6.99E-02	7.70E-03	3.66E-01	2.57E-02	4.70E-01
Sr-90	1.65E-02	--	2.95E+01	--	2.95E+01
Pu-239	2.41E-03	2.49E-04	1.61E+01	3.81E+00	1.99E+01
Pu-238	6.70E-02	8.07E-03	2.39E+01	4.44E-03	2.39E+01
Cm-244	1.09E-03	4.33E-05	--	1.70E-04	1.31E-03
Np-237	4.26E-04	--	--	7.70E-03	8.14E-03
Am-241	3.28E-01	4.22E-01	--	3.19E+00	3.96E+00

CONCLUSIONS

To put the computed estimate of radionuclide activities within LTR IOU into context, attention is drawn to Cs-137, which is the radionuclide with the highest activity levels within LTR IOU Sediment and Sed/Soil materials, and which is the radionuclide that is the biggest contributor to the calculated dose to a member of the public at the perimeter of SRS.

Over the course of the active mission of SRS, P- and R-Reactor secondary cooling water and R-Reactor Disassembly Basin purge water discharges were the three sources of Cs-137 released into the LTR canal system, Par Pond and lower LTR reaches. Based on the findings documented in [1], the secondary cooling water Cs-137 activity releases from P- and R-Reactors are estimated to be 4.4E-04 GBq. This is minor in comparison to the R-Reactor Disassembly Basin releases, which is undoubtedly the primary source of the Cs-137 found in the LTR IOU today. Information in the same report indicates that slightly more than 8.21E+03 GBq of Cs-137 was released to LTR/Par Pond from R-Reactor Disassembly Basin (estimate based upon concentration and discharge volume measurements). Allowing for a radioactive decay correction to 2012, this indicates that approximately 2.59E+03 GBq of Cs-137 would remain in LTR IOU.

A 1991 estimate of the Cs-137 inventory within the LTR/PAR Pond system was made based upon the 8.21E+03 GBq Cs-137 released to the system, radioactive decay, and transport out of the system to the Savannah River [4]. This estimate resulted in a total system Cs-137 inventory of 3.07E+03 GBq with 2.12E+03 GBq above PAR Pond dam and 9.55E+02 GBq below. Decay correcting this to 2012 without accounting for export out of the system results in a total system Cs-137 inventory of 1.89E+03 GBq with 1.31E+03 GBq above the PAR Pond dam and 5.88E+02 GBq below the dam.

A third estimate of Cs-137 activity within LTR IOU is documented in the ERD 2001 report, which was basis for the LTR IOU Cs-137 inventory cited in the [3] report. That report indicated the 2001 Cs-137 activity in LTR IOU to be 2.38E+04 GBq. Decaying correcting this activity to 2012 indicates that an estimated 1.89E+04 GBq remain in LTR IOU in 2012. This estimate is thought to be relatively uncertain because of the lack of documentation of exactly how the inventory was computed. The estimate exceeds the previously cited total releases of Cs-137 from the P- and R-Reactor secondary cooling water and R-Reactor Disassembly Basin releases to LTR IOU and may have included some of the data previously discussed in Section 2.2 (NaI method of analysis) which introduced a high bias in the analytical results.

Thus, the range of total Cs-137 activity remaining in LTR IOU from the three previous estimates is from 1.89E+03 GBq to 1.89E+04 GBq. The estimate in this investigation determined that 1.83E+03 GBq remain above Par Pond Dam and 8.07E+02 GBq below the Dam for a total of 2.63E+03 GBq Cs-137 within the entire LTR IOU. These numbers are quite similar to, but slightly higher than the estimates documented in [1] and [4].

REFERENCES

[1] Carlton, W. H., Bauer, L. R., Evans, A. G., Geary, L. A., Murphy, C. E., Pinder, J. E., and Strom, R. N. 1992. Cesium in the Savannah River Site Environment (U), WSRC-RP-92-250, Westinghouse Savannah River Company, Aiken, SC. March 1992.

[2] ERD (Environmental Restoration Division). 2001. Environmental Restoration Division Response to the Sitewide EM Risk/Hazard Analysis, Environmental Restoration Division Internal Memorandum from W. A. Wilson to T. F. Gaughan, ERD-2001-00950, Westinghouse Savannah River Company, Aiken, SC.

[3] Hiergesell, R. A. M. A. Phifer, J. R. Cook, K. E. Young, M. B. Birk, W. B. Dean. 2008. Inventory of Residual Radioactive Material at the Projected Savannah River Site End State. SRNL-STI-2008-00380, Rev. 0. Savannah River Nuclear Solutions. Aiken, SC 29808.

[4] Marter, W. L. and Boni, A. L. 1991. Summary of Radiological Impact of PAR Pond Drawdown, SRNL-ETS-91-0579, Westinghouse Savannah River Company, Aiken, SC.

[5] SRNS 2009. Periodic Report 3 for the Lower Three Runs Integrator Operable Unit (U), SRNS-RP-2009-00763, Rev. 0, Savannah River Nuclear Solutions, Aiken, SC. May 2009.

[6] SRNS 2012. Periodic Report 4 for the Lower Three Runs Integrator Operable Unit (U), SRNS-RP-2011-01535, Rev. 0, Savannah River Nuclear Solutions. Aiken, SC. March 2012.

[7] WSRC 2002. RCRA Facility Investigation/Remedial Investigation/Remedial Investigation Report with Baseline Risk Assessment for the R-Reactor Seepage Basins/108-4R Overflow Basin Operable Unit (U), WSRC-RP-98-00314, Rev. 1.2, Westinghouse Savannah River Company. Aiken, SC. February 2002

[8] WSRC 2004. Periodic Report for the Lower Three Runs Integrator Operable Unit (U), WSRC-RP-2003-4151, Rev. 0, Westinghouse Savannah River Company. Aiken, SC. June 2004.

[9] WSRC 2006. Periodic Report 2 for the Lower Three Runs Integrator Operable Unit (U), WSRC-RP-2006-4016, Rev. 0, Washington Savannah River Company. Aiken, SC. March 2007.