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# Computation of the Residual Radionuclide Activity Within Three Natural Waterways at the Savannah River Site - 14136

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## ABSTRACT

In 2010 a Composite Analysis (CA) of the U.S. Department of Energy's (DOE's) Savannah River Site (SRS) was completed [1]. This investigation evaluated the dose impact of the anticipated SRS End State residual sources of radionuclides to offsite members of the public. Doses were assessed at the locations where SRS site streams discharge into the Savannah River at the perimeter of the SRS. Although the model developed to perform this computation indicated that the dose constraint of 0.3 mSv/yr (30 mrem/yr), associated with CA, was not approached at the Points of Assessment (POAs), a significant contribution to the total computed dose was derived from the radionuclides (primarily Cs-137) bound-up in the soil and sediment of the drainage corridors of several SRS streams.

DOE's Low Level Waste Federal Review Group (LFRG) reviewed the 2010 CA and identified several items to be addressed in the SRS Maintenance Program. One of the items recognized Cs-137 in the Lower Three Runs (LTR) Integrator Operable Unit (IOU), as a significant CA dose driver. The item made the recommendation that SRS update the estimated radionuclide inventory, including Cs-137, in the LTR IOU. That initial work has been completed and its radionuclide inventory refined [2]. There are five additional streams at SRS and the next phase of the response to the LFRG concern was to obtain a more accurate inventory and distribution of radionuclides in three of those streams, Fourmile Branch (FMB), Pen Branch (PB) and Steel Creek (SC). Each of these streams is designated as an IOU, which are defined for the purpose of this investigation as the surface water bodies and associated wetlands, including the channel sediment, floodplain sed/soil, and related biota. If present, radionuclides associated with IOUs are adsorbed to the streambed sediment and soils of the shallow floodplains that lie immediately adjacent to stream channels.

The scope of this effort included the evaluation of any previous sampling and analysis data that had been collected for various SRS investigations, as well as the additional streambed and floodplain sampling and analysis data acquired more recently as part of the ongoing SRS IOU program, and associated specifically with the FMB, PB, and SC IOUs. Samples have been acquired along the waterways, within the stream channels themselves and in the adjacent floodplain zones. While Cs-137 is the most significant and abundant radionuclide associated with the SRS waterways, it is not the only radionuclide, hence work was conducted to evaluate all radionuclides present.

## INTRODUCTION

FMB, PB, and SC IOUs are located adjacent to one another in the south-central portion of SRS. The watershed boundary associated with each IOU is shown by the areas outlined in red in Figure 1. Each watershed contains facilities and outfalls, as well as waste units in various stages of the remediation process that may represent a potential impact to the associated IOU depending on groundwater and surface water flow paths. Their headwaters occur in the central part of SRS and they flow southwestward to where each discharges into the Savannah River. Various SRS operating facilities are located near the headwaters or mid-sections of these streams and are the source of various discharges which contributed to the buildup of residual radioactivity in stream sediments.

The scope of this effort included the evaluation of sampling and analysis data that existed at the time of the previous inventory estimate, as well as the additional streambed and floodplain sampling and analysis data acquired afterwards as part of the ongoing SRS environmental restoration program, and specifically for the FMB, PB, and SC IOUs. While Cs-137 is the most significant and abundant radionuclide associated with the three IOUs, it is not the only radionuclide present; hence the scope includes evaluating all radionuclides present and includes an evaluation of inventory uncertainty for use in sensitivity and uncertainty analyses. The scope also involved evaluation of the radionuclide inventory associated with the sediments of L-Lake, which is an impoundment constructed on SC and is part of SC IOU.


**Figure 1 Location of FMB, PB and SC Watersheds at SRS**

## Description of FMB, PB AND SC Integrator Operable Units

FMB originates near the center of SRS and follows a southwesterly direction for approximately 24 km (15 mi). The watershed drains about 57 $km^2$ (22 $mi^2$) and includes all or parts of several SRS facilities: all of C-Area (C Reactor), virtually all of N-Area (Central Shops), and those parts of F-, H- and E-Areas (General Separations Area) and the Solid Waste Disposal Facility that lie south of the groundwater divide separating

Upper Three Runs (UTR) and FMB. At its headwaters, FMB is a small backwater stream un-impacted by historical SRS operations. FMB has historically and currently receives effluents from F-, H-, and C-Areas, as well as contaminated groundwater discharges that have migrated from SRS facilities and waste units into the stream and its tributaries. [3]. The FMB IOU is defined as FMB and its tributaries, including surface water and groundwater seeps, sediment (below water surface), sediment/soil (floodplain soil), and related biota in these streams and their associated floodplains. This area represents integration of potential contaminant exposure pathways to on-site human and ecological receptors from potential SRS contaminant sources. The mouth of FMB, where it discharges into the Savannah River (SR) is also the exposure point for offsite members of the public.

The FMB IOU has been sub-divided into three principal components, as indicated in Figure 2. The FMB upper compartment delineates a segment of FMB that is upgradient from process facilities and waste areas and is unlikely to have any SRS process discharges or runoff. In addition, this compartment likely has a narrower and less developed channel and floodplain than the middle and lower reaches of FMB.

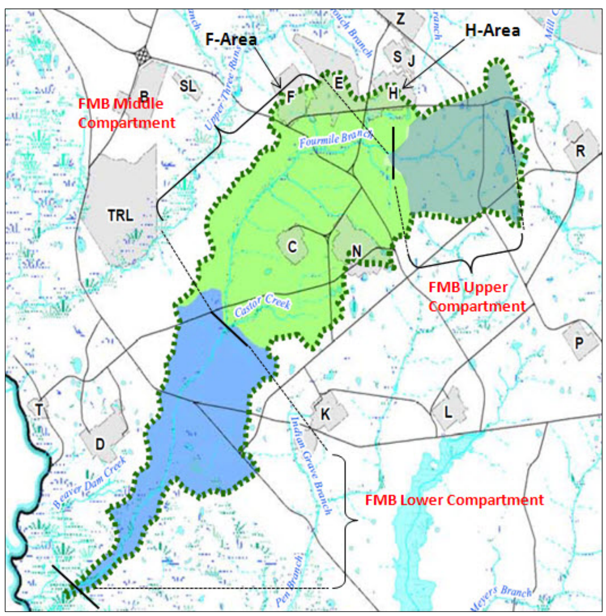


**Fig. 2 Delineation of Fourmile Branch Watershed Compartments**

The upper portion of the FMB middle compartment received discharges from F- and H-Areas including cooling water, process and sanitary wastewater and runoff from waste units, various spills and releases. Shallow groundwater contaminated with metals, organics and radionuclides discharges at seep locations and adds to the material that enters FMB. Large volumes of water were discharged at high flow rates from C-reactor, located downstream from F- and H-Areas, resulting in transport and deposition of sediment contaminated by radionuclides, metals, and organics along the downstream lengths of FMB. Screening data indicate that Cs-137 is present in contaminated media at this locality with the highest activities located within and adjacent to the FMB channel. In general, these data indicate that Cs-137 activity is greatest in the FMB channel and floodplain sediment downstream from the General Separations Areas (GSA), in the vicinity of F-Area. The data indicate a trend of generally decreasing activity in the downstream direction and a trend of decreasing activity with depth, suggesting that Cs-137 activity is associated with near-surface sediments. The lower portion of the FMB Middle compartment includes Castor Creek, which is situated on the east side of FMB. This portion of FMB received discharges of C-Area Reactor cooling water during its period of operation, resulting in a widening of the original channel and deposition of contaminated sediments in floodplain areas along the portion of FMB adjacent to C-Area and extending to the mouth of FMB. Reduction of flow rates following the cessation of reactor operations resulted in retreat of the stream, leaving potentially contaminated areas within the floodplain exposed.

In addition, within the FMB middle compartment, four additional sub-compartments were broken out for this analysis. These include two areas between the F-Area seepage basins and FMB where a grid-like sed/soil sampling program was conducted and an area between the H-Area seepage basins where a similar sed/soil sampling program was undertaken. These two areas are shown in Figure3, with the sample locations indicated and a light-blue box, which indicates the size of the region used to calculate the volume and mass of contaminated soil.

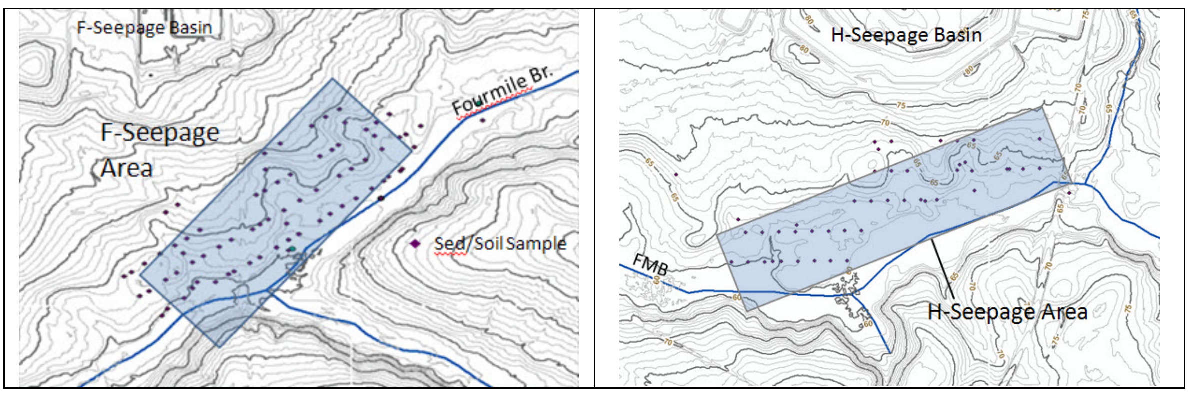


**Fig. 3 Location of the F- and H- Seepage areas**

Two FMB tributary streams, one originating near F-Area and the other near H-Tank Farm, were also broken out separately because of the potential for contaminated material to exist along their reaches and the presence of sediment and sed/soil samples to characterize the amount of radionuclides present. Both of these, which discharge into FMB, are illustrated in Figure 4. The streams themselves are indicated with the pale blue, transparent overlay. Also indicated are the locations of Sediment samples (green dots) and sed/soil samples (purple diamonds). The clusters of sed/soil samples at the upper reach of the H-Area tributary reflect a sampling program to characterize contaminated soil associated with Warner's Pond. These soils have been removed and disposed of within the General Separations Consolidated Unit (i.e. the Old Radioactive Waste Burial Grounds) and are consequently not considered as part of the FMB IOU.

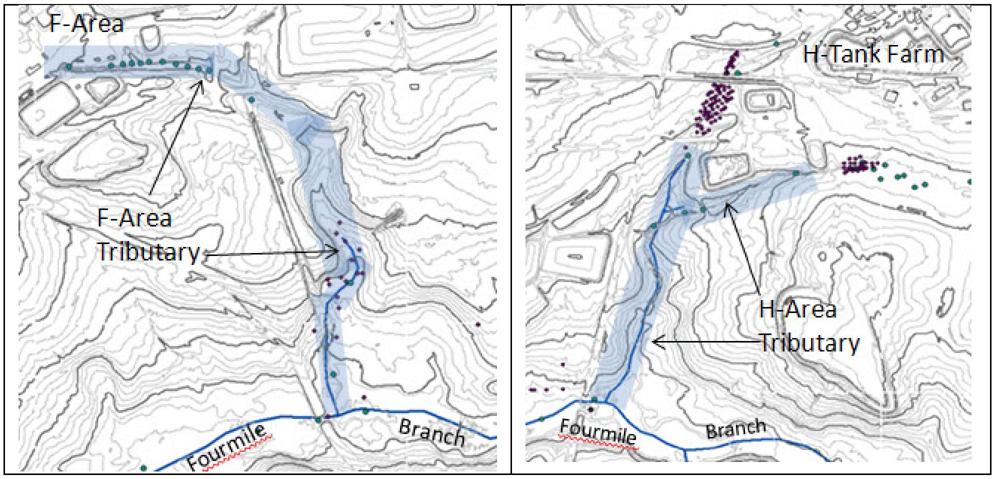


**Figure 4 Location of F-Area and H-Tank Farm tributaries to FMB s**

The conceptual site model (CSM) for the PB IOU divides the watershed into four principal compartments, PB upper, PB middle, Indian Grave Branch, and PB lower, as indicated in Figure 5. The PB upper compartment delineates a segment of PB that is upgradient from process facilities and waste areas and is unlikely to have any SRS process discharges or runoff. In addition, this compartment likely has a narrower and less developed channel and floodplain than the milled and lower reaches of PB.

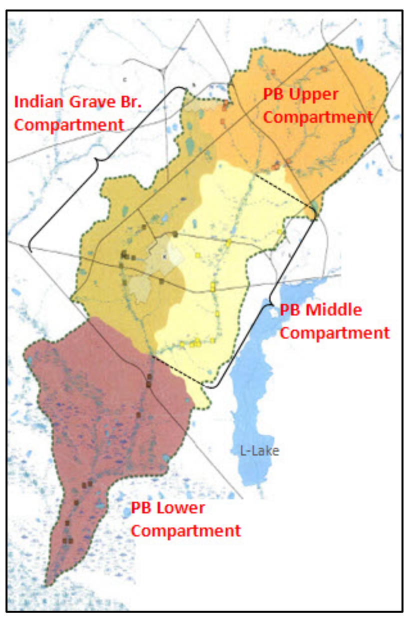


**Figure 5 Delineation of Pen Branch Watershed Compartments**

The PB Middle compartment, which extends from the PB Upper compartment to the confluence with the Indian Grave Branch, may have received contaminated surface runoff in the vicinity of the L-Area ash basin and from surface water runoff from certain operations and from outfalls in the vicinity of K-Area [4].

The Indian Grave Branch compartment is located roughly parallel to the PB Middle compartment, both of which surround K-Area. Overall, Indian Grave Branch received potentially contaminated discharges from its upper reaches all the way to its confluence with PB. These include contributions from outfalls, process water from the K-Area discharge canal and surface runoff from the ash basin [4]. Indian Grave Branch and the PB lower compartment received higher volumes of K-Reactor cooling water discharge during the reactor operations period resulting in a widening of the original channel and deposition of contaminated sediments in floodplain areas along these reaches. Reduction of flow rates following the cessation of reactor operations resulted in retreat of the stream, leaving potentially contaminated areas within the floodplain exposed.

The PB Lower compartment extends from just below the confluence of Indian Grave Branch and PB to the SR. This reach did not receive direct contaminated surface runoff or groundwater discharge from SRS operations, however it is a corridor through which contaminated surface water and sediment migration from upstream has occurred. Additional information can be viewed in the different Periodic Reports for PB [5] [6] [7]

The SC IOU has been sub-divided into four principal compartments, as indicated in Figure 2-6. The SC upper compartment delineates a segment of SC that extends from a location adjacent to P-Area to where SC flows into L-Lake. This segment of SC received reactor cooling water, process water and contaminated effluent from various releases and spills. The large volumes of water discharged at high flow rates resulted in contaminated sediments beyond the creek channel and into the flood plain [8].

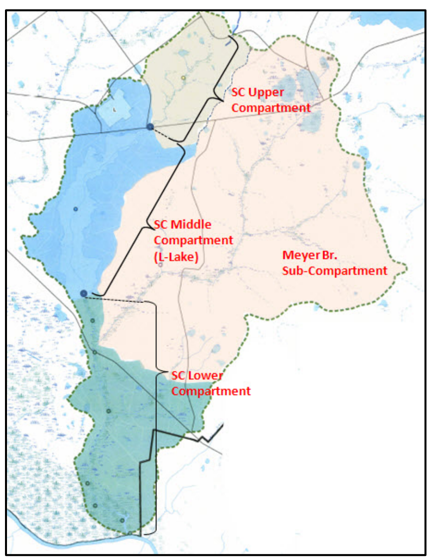


**Figure 6 Delineation of Steel Creek Watershed Compartments**

The SC middle compartment includes L-Lake from where SC enters the upstream end of the lake to L-Lake dam. L-Lake received process and cooling water discharges from P-Reactor via SC and discharges from L-Area units via the L-Area diversion canal.

The SC lower compartment extends from the L-Lake dam to the SR, including the SC delta and swampy lowland adjacent to the SR and extending to the SRS boundary. In this investigation, the lower compartment extends downstream approximately 4.2 km from the confluence of SC and Meyers Branch to the SC delta. The SC delta is where SC enters the SR swamp, which forms the transition between the SC delta and the SR, prior to discharging into the SR. The SC delta and SR floodplain will be evaluated at a later date.

Prior to construction of the L-Lake dam in 1985, the creek channel for lower SC received discharges directly from the reactor areas. Higher discharge rates and volumes resulted in a widening of the original channel and deposition of contaminated sediments in floodplain areas. L-Lake was created in 1985, and the reactors were shut down in 1988, decreasing water releases to SC significantly. The reduced flow resulted in retreat of the stream, leaving potentially contaminated areas within the floodplain exposed [8]. Additional information can be viewed in the different Periodic Reports for SC [9], [10], [11].

## METHOD

The overall approach to computing radionuclide inventories for each of the IOUs involved the following components:

- Defining contaminated reaches of sediments along the IOU waterways
- Identifying separate segments within each IOU waterway to evaluate individually
- Computing the volume and mass of contaminated soil associated with each segment, or “compartment”
- Obtaining the soil core analytical results associated with the channel and floodplain of each IOU compartment
- Computing representative concentrations for all radionuclides associated with the contaminated soil associated with each compartment in each of the IOUs
- Decay correcting analytical results to today, for all sample results
- Computing the radionuclide inventories for each compartments of each IOU by applying the representative concentration to the mass of contaminated soil
- Totaling the inventory for all compartments associated with each of the IOUs

The first step was to segregate sediment and sed/soil analytical results by IOU compartment within the Geographic Information System (GIS) program ArcMap and then export those data for processing. The data for a particular compartment was further segregated by individual radionuclide within separate worksheets so that the estimate of a central, representative value for sediment or sed/soil activity concentration (activity/kg) could be computed.

The radionuclides of interest, for which analyses were performed and whose presence could be confirmed in at least one compartment or sub-compartment of the three IOUs, include the following: Am-241, Am-243, C-14, Cs-137, Co-60, Cm-244, Eu-154, I-129, Np-237, Ni-63, Pu-238, Pu-239, Pu-242, Sr-90, Tc-99, Th-230, U-234, U-235, U-238.

**Issues related to the use of sample analysis data**

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 data analytical qualifiers associated with individual analyses. The qualifiers /connotations associated with the data evaluated in this report include the following:

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

J – species/isotope analyzed for and detected, but the 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. Negative results are qualified as non-detected values (U qualified).

For a group of analysis results associated with samples collected in a particular compartment (or sub-compartment) of one of the IOUs, the following rules were applied:

- If the presence of radionuclide is confirmed in any single 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 compartment/sub-compartment 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.

The rules and logic for selecting data to include in the calculation of the central tendency for Sediment and Sed/Soil samples in individual compartments are based upon the protocol described in [12].

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, 2013. 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 SRS IOUs is not the result of direct release of that radionuclide from DOE operations, but rather occurs as in-growth 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.

The presence of U-234, U-235 and U-238 in the FMB, PB and SC IOUs are a mixture of natural uranium and DOE-added uranium but are dominated by natural uranium. Using the 2X R-Area uranium background values, the sediment and sed/soil representative central values computed for each IOU compartment were compared to see if they exceeded these concentration values 0.057 Bq/g (1.55 pCi/g) for U-233/234, 3 mBq/g (0.078 pCi/g) for U-235 and 0.052 Bq/g (1.4 pCi/g) for U-238; [13] and thus indicated whether DOE-added uranium was present. The majority of FMB, PB and SC compartments were all shown to have concentrations less than these background numbers. Only FMB and SC IOUs contained any DOE-added uranium, this occurring in the SC Upper compartment and in the FMB Middle compartment. More specifically, in FMB, the DOE added uranium occurred in the Middle compartment channel sediment, the F-Area tributary sediment, the Middle compartment sed/soil and the F-Area Seepage zone sed/soil. The DOE-added inventory was computed by subtracting the R-Area background isotope concentration from the computed representative central value of the same isotope for each of these compartments and converting that into an inventory.

Similar to the uranium isotopes, Th-230 also occurs naturally and therefore is not considered to be present as DOE added in FMB, PB and SC IOU compartments unless the representative central value was determined to be greater than a threshold of 0.052 Bq/g (1.4 pCi/g). Again, Th-230 presence was dominated by the natural form and found to be present as DOE-added only in FMB Middle sed/soil and in the SC Upper compartment sediment and sed/soil

## Estimating the Mass of Contaminated Sediment and Sed/Soil

Once a representative activity level was determined for contaminated sediment and sed/soil associated with each group of analytical results associated with each of the FMB, PB and SC IOU compartment/sub-compartment, 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 sediment 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.

With respect to selecting a nominal, representative depth for use in determining the volume of contaminated material, a major issue had to be addressed. The issue is that the sediment and sed/soil sample data recorded in the SRS database suffers from a pervasive lack of mention of a depth intervals associated with the samples. This issue was carefully evaluated in the investigation described in [2], where sufficient depth information was present to make reasonable assumptions for determining a valid nominal and representative depth for each sample type.

The issue is not as significant for sediment samples, because there is ample evidence from previous programs conducted at the SRS to indicate that virtually all of the radionuclide activity is contained within the 0-0.2 m depth interval in pond sediment and in the sediment of flowing streams. In this investigation of FMB, PB and SC IOUs, an assumed 0-0.2 m sampling interval for all sediment samples is adopted based on that evidence.

With regard to sed/soil samples, usually acquired in the floodplain region adjacent to the main channels of SRS stream, the data collected in Lower Three Runs (LTR) having associated depth intervals provided a basis to make reasonable assumptions. Many of those samples were obtained from an interval of 0-0.3 m and indicate that the majority of the radionuclide activity is within this interval.  Samples collected from deeper intervals (0.3-1.2m) indicated that some radionuclide activity can occur in the deeper zone at much lower concentrations but that the greatest concentration resides within the 0-0.3 interval. To account for the small amount of activity associated with the deeper 0.3-1.2m. interval, a nominal depth of contaminated material of 0.35 m was used as a representative depth in the estimate of contaminated soil volumes and mass for all three IOUs.

## 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 FMB, PB and SC IOUs**

| Nuclide | FMB IOU (GBq) | PB IOU (GBq) | SC IOU (GBq) |
|---|---|---|---|
| Am-241 | 3.56E+01 | -- | 9.07E-03 |
| Am-243 | 7.03E+00 | -- | -- |
| C-14 | 2.99E-02 | -- | 1.04E-01 |
| Cs-137 | 3.38E+02 | 5.55E+01 | 6.44E+02 |
| Co-60 | 3.46E+00 | 2.25E+00 | 2.61E+00 |
| Cm-244 | 3.19E+01 | 6.25E-02 | 3.18E-01 |
| Eu-154 | 6.14E-01 | -- | -- |
| I-129 | 1.67E+02 | 4.55E+00 | -- |
| Np-237 | 2.31E+00 | -- | 9.32E+00 |
| Ni-63 | 8.29E+00 | -- | 6.92E+00 |
| Pu-238 | 7.29E+00 | 1.42E-01 | 2.23E+00 |
| Pu-239 | 2.67E+00 | 6.14E-01 | 1.97E+00 |
| Pu-242 | -- | 7.44E+01 | -- |
| Sr-90 | 1.17E+02 | 6.99E-01 | 1.39E+01 |
| Tc-99 | 1.28E+02 | 3.60E+00 | 2.44E-01 |
| Th-230 | 1.42E+00 | -- | 3.58E+00 |
| U-234 | 3.96E+02 | -- | 1.49E+00 |
| U-235 | 4.51E+01 | -- | 1.11E-01 |
| U-238 | 5.33E+02 | -- | 1.48E+00 |

## CONCLUSIONS

This investigation is the first at SRS to attempt to quantify the residual activity of all detectable radionuclides in SRS streams. Since the only previous estimates of any radionuclide were for Cs-137, attention is drawn to that radionuclide to put the computed estimate of radionuclide activities within FMB, PB and SC IOUs into context. Cs-137 is also the radionuclide with the largest dose contribution to the calculated dose to a member of the public at the perimeter of SRS, as documented within the 2010 SRS CA [1].

Over the course of the active mission of SRS, C-, L- and P-Reactor secondary cooling water and Disassembly Basin purge water discharges were sources of Cs-137 released into FMB, PB and SC reaches, respectively. Estimates have been made of the total releases of Cs-137 from each of these reactors into the SRS streams which, when decayed to 2013, provide an upper bounding limit for any independently derived estimate of residual Cs-137 that might remain within the stream corridor into which the releases were made. These releases are 1.03E+03 GBq (27.8 Ci), 2.88E+02 GBq (7.78 Ci) and 3.55E+03 GBq (95.9 Ci) for FMB, PB and SC IOUs, respectively.

The current estimates of residual Cs-137 for each of these IOUs is 3.38E+02 GBq (9.13 Ci), 5.55E+01 GBq (1.5 Ci) and 6.44E+02 GBq (17.4 Ci), respectively, which represent fractions of the total release of 0.33, 0.19 and 0.18. Considering that Cs-137 was released in conjunction with high discharge rates of reactor cooling water at the headwater locations of the streams, the relatively low percentages of Cs-137 actually adsorbing to the stream channel sediments are not surprising. Additionally, since the releases took place over 15 to 20 time-periods, channel sediment with adsorbed Cs-137 was subject to export from the system by ongoing migration of that sediment along the streambed as well as to normal radioactive decay.

While previous estimates of Cs-137 residual activity have been made in FMB, PB and SC, the calculation method was either not documented or the estimated +/- of the estimate was rather large. One prior estimate in SC was well documented and was based on collecting soil cores along the SC floodplain and channel and performing laboratory analyses [14]. These soil cores were collected and analyzed in 1981, providing the basis for a dependable estimate of residual Cs-137 in SC reaches. When decayed to 2013, this earlier estimate for SC conforms closely with the current inventory estimate, which was also based on soil core data, albeit much more recently acquired data. The earlier estimate yielded a residual activity for Cs-137 of 7.81E+02 GBq (21.1 Ci), when decayed to the present, which is highly similar to the current estimate of 6.44E+02 GBq (17.4 Ci) and lends a measure of credibility to the calculations performed in this investigation.

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