Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

Disclaimer:

This work was prepared under an agreement with and funded by the U.S. Government. Neither the U.S. Government or its employees, nor any of its contractors, subcontractors or their employees, makes any express or implied:

- 1) warranty or assumes any legal liability for the accuracy, completeness, or for the use or results of such use of any information, product, or process disclosed; or
- 2) representation that such use or results of such use would not infringe privately owned rights; or
- 3) endorsement or recommendation of any specifically identified commercial product, process, or service.

Any views and opinions of authors expressed in this work do not necessarily state or reflect those of the United States Government, or its contractors, or subcontractors.



Radionuclide Inventories for the F- and H-Area Seepage Basin Groundwater Plumes

Robert A. Hiergesell Walter P. Kubilius

May 2016 SRNL-STI-2016-00273, Revision 0

SRNL.DOE.GOV

DISCLAIMER

This work was prepared under an agreement with and funded by the U.S. Government. Neither the U.S. Government or its employees, nor any of its contractors, subcontractors or their employees, makes any express or implied:

1. warranty or assumes any legal liability for the accuracy, completeness, or for the use or results of such use of any information, product, or process disclosed; or

2. representation that such use or results of such use would not infringe privately owned rights; or

3. endorsement or recommendation of any specifically identified commercial product, process, or service

Any views and opinions of authors expressed in this work do not necessarily state or reflect those of the United States Government, or its contractors, or subcontractors.

Printed in the United States of America

Prepared for U.S. Department of Energy

Keywords: : *Inventory, FMB, PB, SC IOU* **Retention:** *Permanent*

Radionuclide Inventories for the F- and H-Seepage Basin Groundwater Plumes

Robert A. Hiergesell Walter P. Kubilius

May 2016



Prepared for the U.S. Department of Energy under contract number DE-AC09-08SR22470.

OPERATED BY SAVANNAH RIVER NUCLEAR SOLUTIONS

REVIEWS AND APPROVALS

AUTHORS:

Robert A. Hiergesell, Environmental Modeling, SRNL	Date
Walter P. Kubilius, Engineering Process Development, SRNL	Date
TECHNICAL REVIEW: (Reviewed per E7 2.60)	
Thong Hang, Environmental Modeling, SRNI	Date
APPROVAL:	Date
David A. Crowley, Manager Environmental Modeling, SRNL	Date
Kevin M. Kostelnik, Director Environmental Restoration Technologies, SRNL	Date

EXECUTIVE SUMMARY

Within the General Separations Areas (GSA) at the Savannah River Site (SRS), significant inventories of radionuclides exist within two major groundwater contamination plumes that are emanating from the F- and H-Area seepage basins. These radionuclides are moving slowly with groundwater migration, albeit more slowly due to interaction with the soil and aquifer matrix material. The purpose of this investigation is to quantify the activity of radionuclides associated with the pore water component of the groundwater plumes.

The scope of this effort included evaluation of all groundwater sample analyses obtained from the wells that have been established by the Environmental Compliance & Area Completion Projects (EC&ACP) Department at SRS to monitor groundwater contamination emanating from the F- and H-Area Seepage Basins. Using this data, generalized groundwater plume maps for the radionuclides that occur in elevated concentrations (Am-241, Cm-243/244, Cs-137, I-129, Ni-63, Ra-226/228, Sr-90, Tc-99, U-233/234, U-235 and U-238) were generated and utilized to calculate both the volume of contaminated groundwater and the representative concentration of each radionuclide associated with different plume concentration zones.

The overall approach to computing radionuclide inventories for each of the seepage basin groundwater contaminant plumes involved the following:

- Evaluating groundwater sample analytical results obtained from wells surrounding F- and H-Area seepage basins
- Defining the likely extent of groundwater contaminant plumes emanating from the seepage basins
- Defining concentric zones of contaminant concentration ranges within each groundwater plume within each of the three shallow aquifer units for each radionuclide
- Estimating the volume of the groundwater contained within each contaminant plume zone
- Determining a representative pore water concentration to associate with each contaminant plume zone

The total calculated activity for each radionuclide is presented below for the F- and H-Area groundwater contamination plumes.

	F-Area Groundwater Plume	H-Area Groundwater Plume
Nuclide	(Ci)	(Ci)
Am-241	4.59E-03	5.27E-05
Cm-243/244	1.77E-03	1.61E-05
Cs-137	7.99E-02	0.00E+00
I-129	2.01E-01	8.84E-03
Ni-63	0.00E+00	2.04E-02
Ra-226/228	8.15E-02	6.63E-03
Sr-90	4.78E-01	6.45E-02
Tc-99	2.25E-01	1.81E-01
U-233/234	8.75E-02	1.53E-03
U-235	1.10E-02	7.51E-05
U-238	2.44E-01	1.66E-03

The estimated total uncertainty of these activities is approximately $\pm 80\%$ for all radionuclides.

TABLE OF CONTENTS

LIST OF FIGURES vii
1.0 Introduction 1
1.1 Purpose and Scope
1.2 Description of F- and H-Area Seepage Basins and General History
2.0 Method
2.1 Groundwater Sampling and Data Processing
2.2 Use of ArcMap to Facilitate Analysis
2.3 Areal Extent of Contaminant Plumes
2.4 Volume of Groundwater Plume Zones
3.0 Results
3.1 Evaluation of Uncertainty in Inventory Estimate
3.2 Contaminants Sorbed to Formation
4.0 References
APPENDIX A 15
Groundwater Plume Maps for H-Area Seepage Basin15
APPENDIX B
Groundwater Plume Maps for F-Area Seepage Basin27

LIST OF TABLES

Table 3-1	Pore Water Activity Levels for F- and H-Area Seepage Basins	9
Table 3-2	Cumulative and Decay-Corrected Discharges to F- and H-Area Seepage Basins	9
Table 3-3	Total Calculated Pore Water Activity from all Aquifer Units 1	0
Table 3-4	Total Uncertainty Associated with Groundwater Plume Radionuclide Estimates 1	2

LIST OF FIGURES

Figure 1-1	Location of F- and H-Area Seepage Basins at SRS	3
Figure 2-1	Cross-Section utilized to Plot Radionuclide Plume Thicknesses in F-Area	6
Figure 2-2	Cross-Section utilized to Plot Radionuclide Plume Thicknesses in H-Area	7
Figure 2-3	Methods of Groundwater Plume Zone Volume Calculation	7

LIST OF ACRONYMS

ArcMap	Geographic Information System Program
CA	Composite Analysis
DOE	Department of Energy
EC&ACP	Environmental Compliance & Area Completion Projects
EMS	Environmental Monitoring Section
EPA	Environmenatl Protection Agency
ERDMS	Environmental Restoration Data Management System
FSB	F-Area Seepage Basin
GA	Gordon Aquifer
GSA	General Separations Area
HSB	H-Area Seepage Basin
HWMF	Hazardous Waste Management Facility
K _d	Contaminant Partitioning Coefficient
LAZ	Lower Aquifer Zone of the Upper Three Runs Aquifer
LFRG	Low Level Waste Disposal Facility Federal Review Group
MCL	Maximum Contaminant Level
OU	Operable Unit
ORWBG	Old Radioactive Waste Burial Ground
POA	Point of Assessment
RCRA	Resource Conservation and Recovery Act
SRNL	Savannah River National Laboratory
SRNS	Savannah River Nuclear Solutions
SRS	Savannah River Site
TCCZ	Tan Clay Confining Zone
UAZ	Upper Aquifer Zone of the Upper Three Runs Aquifer
UTM	Universal Transverse Mercator
WSRC	Westinghouse or Washington Savannah River Company

1.0 Introduction

In 2010 the Savannah River National Laboratory (SRNL) completed a Composite Analysis (CA) of the U.S. Department of Energy's (DOE's) Savannah River Site (SRS). That investigation evaluated the dose impact of the anticipated SRS End State residual sources of radionuclides to offsite members of the public. These doses were evaluated at several points of assessment (POAs) located where SRS site streams discharge into the Savannah River (SR). Evaluations were conducted using models developed to perform this computation. The results indicated that the dose constraint (30 mrem/yr) associated with the CA would not be approached at any of the POAs.

DOE provided conditional approval of the SRS CA (SRNL 2010) on July 16, 2010 (Marcinowski 2010). Approval was provided with the condition that the secondary issue identified by the Low Level Waste Disposal Facility Federal Review Group (LFRG) review team (Carilli and Golian 2010) be resolved. The secondary issue identified by the LFRG review team consisted of the consolidation of eighteen observations that the team concluded, when evaluated collectively, could potentially impact the integration of the CA results. Nine of these secondary issue observations, which involved missing information, were resolved by additions to the CA prior to its approval. Specific future work items were added to the CA maintenance plan to provide a path forward for the resolution of the other nine secondary issue observations. Amongst these observations, Observation 2, Item 8 (SRNL 2010 Table 11-2), which identifies the need to quantify the inventory and inventory distribution associated with radionuclides contained within the F- and H-Area Seepage Basin groundwater plumes, is the basis for this investigation.

1.1 Purpose and Scope

Significant inventories of radionuclides exist within two major groundwater contamination plumes that are emanating from the F- and H-Area seepage basins. These radionuclides are moving slowly with groundwater migration, albeit more slowly due to interaction with the soil and aquifer matrix material. The purpose of this investigation is to quantify the activity of radionuclides associated with the pore water component of the groundwater plumes.

The scope of this effort included evaluation of all groundwater sample analyses obtained from the wells that have been established by the Environmental Compliance & Area Completion Projects (EC&ACP) Department at SRS to monitor groundwater contamination emanating from the F- and H-Area Seepage Basins. Using this data, generalized groundwater plume maps for the radionuclides that occur in elevated concentrations (Am-241, Cm-243/244, Cs-137, I-129, Ni-63, Ra-226/228, Sr-90, Tc-99, U-233/234, U-235 and U-238) were generated and utilized to calculate both the volume of contaminated groundwater and the representative concentration of each radionuclide associated with different plume concentration zones. C-14, although present in groundwater in the vicinity of the F- and H-Area seepage basins is not evaluated in this investigation owing to relatively low observed groundwater concentrations (less than half of the Maximum Contaminant Level (MCL) and the difficulty in obtaining accurate analytical results for C-14 when tritium is also present in groundwater samples. When significant tritium activity is present in groundwater samples, the apparent C-14 analytical result can be biased significantly higher than the actual C-14 concentration result.

As this investigation proceeded, it was realized that additional research and characterization efforts are required to adequately quantify the adsorbed species component of the groundwater plume radionuclide inventories, which led to a restriction of the scope of the investigation to quantifying only the pore water component. The need for additional research is primarily due to the strong dependence of the partitioning tendency of different species on groundwater pH conditions, specifically the strongly acidic groundwater pH levels in in the vicinity of the F- and H-Area seepage basins. This issue is discussed more thoroughly in Section 3.2 of this report. Finally, an assessment of the uncertainty associated with the pore water radionuclide inventory calculations was conducted.

1.2 Description of F- and H-Area Seepage Basins and General History

The F-Area Seepage Basins (FSB) (904-41G, -42G, -43G) are part of the F-Area Hazardous Waste Management Facility (HWMF) Operable Unit (OU). The F-Area HWMF OU is located in the central portion of SRS, approximately five miles from the nearest site boundary and operated from 1955 until November 7, 1988. During that time, the facility received waste effluents from F-Area chemical separation facilities processes such as the nitric acid recovery unit, waste storage system evaporator overheads, and general-purpose evaporator overheads. These basins were closed by dewatering, physically and chemically stabilizing the remaining sludge, and placing a protective multi-layer cover system over them to reduce rainwater contact with basin bottoms.

The H-Area Seepage Basins (HSB) are a part of the H-Area HWMF OU, which is located in the central portion of SRS, approximately six miles from the nearest site boundary. The H-Area HWMF operated from 1955 until November 7, 1988. The original H-Area HWMF consisted of Basins 904-44G, 904-45G and 904-46G and operated from 1955 to 1962. In 1962, 904-46G was replaced by 904-56G. At the time of closure, the H-Area HWMF (904-44G, 904-45G, and 904-56G) had a combined maximum operating capacity of 26.5 million gallons of wastewater. Both seepage basins are illustrated in Figure 1-1, with reference to H-Area, the Old Radioactive Waste Burial Ground (ORWBG) and Fourmile Branch.

The FSB and HSB sites are regulated under the Resource Conservation and Recovery Act (RCRA), and groundwater associated with both facilities is undergoing remediation. Groundwater is sampled and analyzed at various times from over 100 monitoring wells at each facility. SRS submits annual Corrective Action Reports to EPA and SCDHEC, most recently in 2015 (SRNS, 2015), with plume maps for I-129, Sr-90, U-238, and tritium. The plume maps are constructed as a "snaphot in time", and therefore include only those wells which were sampled in the third calendar quarter of the previous year.

For this study, it was desired to construct plume maps for all important radionuclides, and to incorporate data from all wells, not just those sampled in a particular quarter. Therefore, the EC&ACP database was queried to supply analytical data for all wells sampled at least once during a three-year period, 2013-2015.



Figure 1-1 Location of F- and H-Area Seepage Basins at SRS

2.0 Method

The overall approach to computing radionuclide inventories for each of the seepage basin groundwater contaminant plumes involved the following:

- Evaluating groundwater sample analytical results obtained from wells surrounding F- and H-Area seepage basins
- Defining the likely extent of groundwater contaminant plumes emanating from the seepage basins
- Defining concentric zones of contaminant concentration ranges within each groundwater plume within the Upper Aquifer Zone (UAZ), Lower Aquifer Zone (LAZ) and Gordon Aquifer (GA) units for each radionuclide
- Estimating the volume of the groundwater contained within each contaminant plume zone
- Determining a representative pore water concentration to associate with each contaminant plume zone
- Calculating the total activity of each radionuclide within the groundwater plumes associated with each seepage basin
- 2.1 Groundwater Sampling and Data Processing

The initial step in the investigation was to obtain all of the available groundwater sample analysis data downgradient from the F- and H-Area seepage basins. This involved identifying the wells impacted by contaminated groundwater emanating from seepage basins. Historical EC&ACP reports were consulted to determine the lateral and vertical extent of groundwater contamination in the shallow subsurface and to identify the wells from which groundwater samples have been obtained to characterize the extent of subsurface contamination. The annual Corrective Action Reports prepared by EC&ACP have been particularly helpful in this effort. One such report is SRNS-RP-2015-00136, Volumes I, II and III.

The radionuclides of interest, for which well sample analyses were performed and whose presence could be confirmed, include the following: Am-241, Cs-137, Cm-243/244, I-129, Ni-63, Ra-226/228, Sr-90, Tc-99, U-233/234, U-235, and U-238.

Several radionuclides were omitted from consideration because there were no credible detections during the study period; these include: Sb-125, Ba-133, Cs-134, Co-60, Cm-242, Cm-245/246, Cs-137 (at HSB), Eu-152, Eu-154, Eu-155, Pu-238, Pu-239/40, Pm-146, and Na-22. Other nuclides were omitted from consideration because they are daughter products of radionculides which are being evaluated; these include Ac-228, Th-228, and Th-230. Ni-63 was omitted for FSB because of insufficient available data. Tritium is present in groundwater at both facilities, but estimation of tritium activity is not included because it was previously addressed in the original CA inventory.

Reported concentrations of two radionuclides are subject to uncertainty due to analytical limitations. Analytical results of C-14 and Ni-63 may be biased high, due to the presence of tritium. Confidence in the accuracy of C-14 analyses is very low, therefore, no plumes were drawn and no activity calculations were performed for this contaminant. However, the highest reported concentration of C-14 during the study period (738 pCi/L) is well below the EPA MCL of 2,000 pCi/L. Therefore, even if there is no high bias in C-14 results due to tritium interference, all groundwater at both FSB and HSB meets the EPA drinking water standard for that nuclide. Ni-63 analytical results may also be affected by tritium interference, and may also have a high bias. However, confidence is greater for Ni-63 than for C-14, so a plume was drawn, and an inventory was calculated for Ni-63 at HSB.

Having identified the relevant wells from which groundwater samples were obtained, historical analytical results for the selected nuclides were downloaded from ERDMS database. Because analytical results are not obtained from every well during each sampling event, and because groundwater sample concentrations can vary from sample event to sample event in a given well, a decision was made to utilize the maximum groundwater concentration result for each radionuclide in each well over the 2013-2015 time period. It is expected that this will introduce bias into the calculated results, but will enable the true radionuclide pore water inventories to be bounded.

2.2 Use of ArcMap to Facilitate Analysis

An ArcMap project was established to develop the groundwater plume maps associated with each of the seepage basins. ArcMap plume layer files for the different radionuclides (by aquifer unit) were created, with tables containing well construction and analytical results. These tables include:

- Well name
- UTM well coordinates
- Well screen zone elevations
- Well reference elevations
- Well aquifer designation (e.g. UAZ, LAZ and GA)
- Maximum concentration for each radionuclide for each well (2013-2015)

An Attribute Table was associated with each radionuclide within each aquifer unit. Individual records in each Attribute Table include several key parameters, including FID (a polygon ID

number) analyte, analyte concentration range, aquifer unit, and the polygon shape area. Images of the ArcMap plume zones associated with each radionuclide, in each aquifer unit near the F- and H-Area seepage basins are presented in APPENDIX A and APPENDIX B.

2.3 Areal Extent of Contaminant Plumes

For each well, the maximum concentration reported during the calendar years 2013-2015 was selected for construction of ArcGIS concentration contour maps. Concentration contour intervals were chosen so that for most radionuclides three concentration zones, Low (L), Medium (M), and High (H), were established. For example, the concentration zones for Sr-90 at FSB are 2 - 8 pCi/L (L), 8 - 100 pCi/L (M), and 100 - 600 pCi/L (H). At each waste unit, plume maps were made for three aquifer zones (UAZ, LAZ, GA). Concentration zones for the UAZ at FSB were further divided spatially into upland and swamp zones, with the boundary between them roughly following the 200-ft land surface elevation contour. This was done in order to account for thinning of the UAZ in the vicinity of swampy lowlands near Fourmile Branch.

Each plume zone on a concentration contour map represents a defined area, within which groundwater contaminant concentrations are postulated to be between a defined lower limit and a defined upper limit (e.g. 8 to 100 pCi/L for the Medium Sr-90 zones at FSB). For each zone, a single representative concentration is assigned to that zone's volume. The representative concentration is the average of all well concentrations for wells located within a particular zone (recall that the well concentrations used are the maximum for a three-year period). For several Low concentration plume zones where there are few wells in a large area, the representative concentration was assigned the midpoint of the zone's concentration limits; for example, the FSB UAZ Sr-90 Swamp Low zone was given a concentration of 5 pCi/L, midway between the limits of 2 and 8 pCi/L.

2.4 Volume of Groundwater Plume Zones

The next work objective was to estimate the volume associated with each radionuclide groundwater plume concentration zone. The first step in this process was to estimate the vertical thickness of the groundwater plumes within each aquifer unit. Geologic cross-section depicting the aquifer units in the vicinity of the F- and H-Area seepage basins were obtained from EC&ACP and are illustrated in Figure 2-1 (F-Area) and Figure 2-2 (H-Area).

Each cross-section extends across the main part of each groundwater contaminant plume at each basin, or that portion of the plume generally containing the highest groundwater radionuclide concentrations. The well screen zones for each of the wells defining the cross-section are illustrated with respect to their position within each aquifer unit, the UAZ, the LAZ or the GA. The uppermost aquifer is the UAZ, the middle aquifer is the LAZ and the lowermost aquifer unit is the GA. The blue line in the upper part of each illustration defines the position of the water table. The two darker horizontal bands represent the confining units that separate the aquifer units. The upper band represents the elevation of the "Tan Clay" and the lower band represents the Gordon Confining Unit (or "Green Clay").

On the cross-sections illustrated in Figure 2-1 and Figure 2-2, the radionuclide 3-year maximum concentrations were plotted by each well screen to create a series of worksheets onto which the vertical configurations of the low, medium and high concentration zones could be sketched. Worksheets were developed for each radionuclide that was detected in groundwater samples

obtained from each aquifer unit for both the F- and H-Area cross-sections. The selected vertical thicknesses for each plume concentration zone, once estimated, were entered into the Attribute Table associated with each ArcMap radionuclide aquifer plume layer in the ArcMap project named "FSB4.mxd". These values were entered into a column within the Attribute Tables with the heading title "report_id"



Figure 2-1 Cross-Section utilized to Plot Radionuclide Plume Thicknesses in F-Area



Figure 2-2 Cross-Section utilized to Plot Radionuclide Plume Thicknesses in H-Area

Calculations of the volume of groundwater plume concentration zones were performed within three MS Excel files, named "UAZ_Plume_Calcs.xlsx", "LAZ_Plume_Calcs.xlsx" and "GA_Plume_Calcs.xlsx". Two methods of estimating the appropriate volume were employed but predominantly a method whereby the plume zones are thought of as concentric zones within one large groundwater plume. This concept is illustrated in Figure 2-3. In this illustration Groundwater Plume Zone 1 is the inner concentration zone and Plume Zone 2 is the outer concentration zone. The total volume of each Plume Zone was calculated by multiplying the area of each of these plume zones by their representative thicknesses. In Figures 2-3, the representative thickness of Plume Zone 1 is the vertical length indicated by "B" while the representative thickness of Plume Zone 2 is vertical length indicated by "A". Because the volume of Groundwater Plume Zone 2 includes the volume of Groundwater Plume Zone 1, the volume of the latter was subtracted from the volume of Groundwater Plume Zone 2 to approximate its actual, or "adjusted", plume volume.



Figure 2-3 Methods of Groundwater Plume Zone Volume Calculation

Within each of the MS Excel spreadsheet tabs associated with a particular radionuclide there are two columns that indicate: 1) Plume Volume and 2) Adjusted Plume Volume (columns I and J). If the Adjusted Plume Volume is different from the Plume Volume this indicates that at least one "inner" concentration zone volume was subtracted from the "outer" plume concentration zone volume. Algorithms indicating how the specific volume subtractions were implemented are illustrated in a box within each tab entitled "Adjustments for concentric Concentration Zones in Aquifers".

The second method for calculating the volume of the "outer" Groundwater Plume Zone was only utilized in a single instance to calculate the inventory of I-129 within the LAZ adjacent to the F-Seepage Basin. For this case the "outer" concentration zone was conceived to be a thin shell of relatively uniform thickness surrounding the "inner" concentration zone. In Figure 2-3, this relatively uniform representative thickness for Groundwater Plume Zone 2 was the vertical length indicated by "C".

Not all groundwater plume concentration zones were located exactly in the vicinity of the hydrogeologic cross-sections (see Figure 2-1). Often, the groundwater plume concentration zones occur as smaller areas of thinner vertical extent than the central portion of the F- and H-Area seepage basin groundwater plumes and are sometimes completely isolated from the centrally located plume zones. Defining a vertical thickness of these zones is difficult due to insufficient

information to accurately define the thickness. Because of this uncertainty, the thickness estimated was based upon an examination of the screen lengths, the vertical position of the well screens within the adjacent aquifer unit, the analytical results and a judgement based on the perceived pattern of groundwater movement.

Having computed the volume of each groundwater plume concentration zone, the fluid water volume was estimated by multiplying this volume by the porosity of each aquifer. A value of 0.38 for sandy material was selected for each aquifer. This value was obtained from the Hydraulic Property Data Package previously developed for use in the SRS E-Area Performance Assessment (Phifer, et.al 2006). A simple multiplication of the representative groundwater concentration with the plume concentration zone fluid volumes yielded the activity level of each radionuclide in each concentration zone in each of the F- and H-Area groundwater contaminant plumes.

3.0 Results

Using the method described in Section 2.0, the calculated groundwater radionuclide inventories for the pore water associated with the F- and H-Area Seepage Basin groundwater plumes were calculated. The total activities for each radionuclide were computed by summing the activity associated with each groundwater plume concentration zone, for each of the seepage basins. These total activities are presented in Tables 3-1 for each of the aquifers, UAZ, LAZ and the GA.

While no accounting is made for adsorbed activity within the groundwater plumes, a useful check is to compare the total pore water radionuclide activity with the total radionuclide activities that have been estimated to have been disposed within each of the seepage basins during the active lives of those facilities. Estimates of disposed activities are available for a subset of the radionuclides in three historical SRS documents (see Killian, et.al. 1985a and Killian, et.al. 1985b and Looney, et.al. 1990). These reports provided an estimate of activities decayed from the time of disposal to a point in time in 1985. The decay corrected data was further decayed to 2016 in this investigation and is presented below in Table 3-2. The activities reported for the combined isotopes of Am-241 and Am-243 are assumed to be entirely attributable to Am-241 in this investigation. Similarly, the activity reported for the combined isotopes of Cm-242 and Cm-244 are assumed to be entirely attributable to Cm-244.

To make a valid comparison for each seepage basin, the pore water activities for each radionuclide were totaled across each of the aquifer units for the F- and H-Area Seepage Basins. This summation is presented in Table 3-3. It is expected that the totaled pore water activity levels

	UAZ		LAZ		GA	
	F-Area	H-Area	F-Area	H-Area	F-Area	H-Area
Nuclide	Basins	Basins	Basins	Basins	Basins	Basins
	(Ci)	(Ci)	(Ci)	(Ci)	(Ci)	(Ci)
Am-241	4.27E-03	5.27E-05	3.22E-04	NA	NA	NA
Cm-243/244	1.55E-03	1.61E-05	2.23E-04	NA	NA	NA
Cs-137	7.99E-02	NA	0.00E+00	NA	NA	NA
I-129	9.74E-02	4.64E-03	1.02E-01	4.19E-03	1.69E-03	8.76E-06
Ni-63	NA	2.97E-03	NA	1.07E-02	NA	6.75E-03
Ra-226/228	4.32E-02	2.22E-03	3.67E-02	3.01E-03	1.57E-03	1.40E-03
Sr-90	1.37E-01	6.39E-02	3.40E-01	5.77E-04	8.78E-04	2.97E-05
Tc-99	4.15E-02	3.30E-02	1.77E-01	1.48E-01	6.43E-03	3.88E-04
U-233/234	8.39E-02	1.53E-03	3.61E-03	NA	NA	NA
U-235	7.35E-03	7.51E-05	3.61E-03	NA	NA	NA
U-238	1.64E-01	1.66E-03	8.03E-02	NA	NA	NA

Table 3-1 Pore Water Activity Levels for F- and H-Area Seepage Basins

would be significantly less than the estimated disposals at each of the basins over the course of their operations. A part of the disposed inventory for the most mobile radionuclides may have already passed entirely through the shallow subsurface and been discharged into Fourmile Branch. Conversely, the least mobile radionuclides may still be adsorbed within the shallow subsurface or be entrained within the treated basin floor as part of the basin closure activities.

	F-Area Seepa	age Basin		H-Area Seepage Basin			
	Cumulative	Decay	Decay	Cumulative	Decay	Decay	
	Release to	Corrected	Corrected	Release to	Corrected	Corrected	
	Basin ¹	to 1985 ¹	to 2016	Basin ²	to 1985^2	to 2016	
	(Ci)	(Ci)	(Ci)	(Ci)	(Ci)	(Ci)	
Am-241	2.16E-01	2.15E-01	2.05E-01	5.07E-02	5.04E-01	4.79E-01	
Cm-244	3.12E-01	2.36E-01	7.20E-02	7.12E-02	5.19E-02	1.58E-02	
Cs-137	2.12E+02	1.35E+02	6.60E+01	1.57E+02	1.12E+02	5.48E+01	
I-129	2.00E+00	2.00E+00	2.00E+00	4.00E-01	4.00E-01	4.00E-01	
Sr-90	4.13E+01	2.31E+01	1.10E+01	4.29E+01	2.69E-01	1.28E-01	
Tc-99	1.00E+00	1.00E+00	1.00E+00	5.00E-01	5.00E-01	5.00E-01	
U-238	-	6.40E-01 ³	6.40E-01	-	-	-	

Table 3-2	Cumulative and	l Decay-Corrected	Discharges to F- and	H-Area Seepage Basins
-----------	----------------	-------------------	-----------------------------	------------------------------

Notes: ¹ From Killian, et.al. 1985a; ² From Killian, et.al. 1985b; ³ From Looney, et.al. 1990

A careful examination of the totaled pore water activity levels associated with the groundwater plumes emanating from each seepage basin reveals they are all lower than the decay-corrected disposal activity levels for the radionuclide data that are reported.

	F-Area	H-Area
	Seepage	Seepage
	Basin Total	Basin Total
	Inventory	Inventory
	from all	from all
Nuclide	Aquifers	Aquifers
	(Ci)	(Ci)
Am-241	4.59E-03	5.27E-05
Cm-244	1.77E-03	1.61E-05
Cs-137	7.99E-02	0.00E+00
I-129	2.01E-01	8.84E-03
Ni-63	0.00E+00	2.04E-02
Ra-226/228	8.15E-02	6.63E-03
Sr-90	4.78E-01	6.45E-02
Tc-99	2.25E-01	1.81E-01
U-234	8.75E-02	1.53E-03
U-235	1.10E-02	7.51E-05
U-238	2.44E-01	1.66E-03

Table 3-3 Total Calculated Pore Water Activity from all Aquifer Units

3.1 Evaluation of Uncertainty in Inventory Estimate

Several different uncertainties affect the estimation of radionuclide activity at the FSB and HSB. For each contaminant, the total activity present in groundwater was calculated as:

activity = (plume area) x (plume thickness) x (porosity) x (contaminant concentration)

Each of the four general variables on the right side of the equation has one or more uncertainties, which are discussed separately below.

Uncertainty in plume area

The overall areal extent of the contaminant plumes are well established at both facilities, due to the outstanding well coverage – over 100 wells at each site – and due to the downgradient end of both plumes being anchored by surface water features. The uncertainty in plume area is estimated as $\pm 10\%$ for plumes in the UAZ and LAZ. The uncertainty in the Gordon Aquifer is larger, because fewer monitoring wells are available. Plume areas estimates in the GA could be off by as much as a factor of two ($\pm 100\%$, -50%), but this uncertainty would have little effect on the overall plume area uncertainty because plume areas and contaminant concentrations are both much smaller in the GA than in the UAZ and LAZ. Therefore, total uncertainty in plume areas, considering all three aquifer zones, is considered to be $\pm 10\%$.

Uncertainty in plume thickness

The maximum possible thicknesses of contaminant plumes are bounded by the thicknesses of the hosting aquifer zones: about 35 feet for the UAZ, 75 feet for the LAZ (plus TCCZ), and 40 feet for the upper half of the GA (the lower half is appears to be uncontaminated). However, most

chosen representative thicknesses of plume polygons are less than the total aquifer zone thickness. The uncertainty in chosen representative plume thicknesses is estimated to be $\pm 50\%$.

Uncertainty in aquifer porosity

The porosity of all three aquifer zones is taken to be 0.38 ± 0.05 , or a 13% uncertainty.

Uncertainty in contaminant concentration

The uncertainty inherent in the estimation of contaminant concentrations is a complicated function, which has several components described below.

Use of maximum detected concentration

In this report, groundwater plumes were constructed by using the maximum concentration of each contaminant in each well during the three calendar years 2013-2015. The maximum was used instead of the average in order to build some conservatism into the inventory estimate. Generally for each contaminant in each well, the maximum reported concentration during 2013-15 is about 40 to 60% higher than the average reported concentration for the same period.

Analytical uncertainty of maximum detected concentration

Each radiological analysis of a contaminant has its own reported 2-sigma counting uncertainty, which can be expressed as a percent uncertainty, relative to the reported concentration. For samples with very low radionuclide concentrations, the percent uncertainty can exceed 50% of the reported result. However, wells with these low concentrations do not contribute much activity to the final inventory calculation. The bulk of the plume's activity resides near wells with higher concentrations. As concentration goes up, the relative percent uncertainty of the concentration goes down, and can be below 10% for samples taken from wells in the core of an analyte's plume. Generally, a representative relative counting uncertainty for samples used in inventory calculations can be assumed to be $\pm 25\%$ at FSB. Concentrations are lower at HSB, so counting uncertainties are estimated to be a bit higher at $\pm 30\%$.

Average concentrations within plume zones

The spatial volume encompassed by a plume zone is assigned a representative concentration, which is determined by averaging the concentrations of sampled wells within that zone (and the well concentrations are the maxima during 2013-2015). A non-parametric Student's t-test evaluation of the 95% Upper Confidence Level of the mean, conducted for the Medium and High contour interval plume zones, suggests that a reasonable estimate of the uncertainty associated with the average concentrations is $\pm 35\%$.

Analyte-specific uncertainty

Radium-226 and radium-228 occur naturally in SRS groundwaters. Therefore, in order to estimate the activity of radium in groundwater, attributable to DOE operations, the natural portion must be estimated, and then subtracted from the measured concentration in every well. Based on the radium concentrations in nearby wells with little or no contamination, the background concentration of (Ra-226 + Ra-228) is estimated to be 2.0 pCi/L in all three aquifer zones. Sensitivity calculations performed by changing the background value to 1.5 and 2.5 pCi/L and recalculating inventory, indicate that the change to the calculated (Ra-226 + Ra-228) inventory would be $\pm 4\%$ at FSB, and $\pm 26\%$ at HSB.

Representativeness of well screen depths

Groundwater contaminant plumes are generally very heterogeneous and spatially complex; this is also true specifically for FSB (Wan et al, 2012). It is not known how many of the groundwater monitoring wells are screened at depths with representative concentrations of contaminants for their aquifer zone. Some wells were intentionally installed to sample the most contaminated level of their aquifer zone; other well screens may have missed the plume. The degree of uncertainty associated with well screen representativeness is not easy to determine, so an arbitrary uncertainty of $\pm 40\%$ is assigned to this variable.

Combined Uncertainty of Plume Activity

Table 3-2 summarizes the uncertainties in calculation of plume activities. Because some of the uncertainties discussed above are subjective, it is not possible to rigorously define a total uncertainty for plume activity. However, we can approximate the total uncertainty in a general fashion by taking the square root of the sum of the squares of individual uncertainties.

Table 3-4	Total	Uncertainty	Associated	with	Groundwater	Plume	Radionu	clide	Estimates
-----------	-------	-------------	------------	------	-------------	-------	---------	-------	-----------

	Percent uncertainty		
	FSB	HSB	
Plume area	10	10	
Plume thickness	50	50	
Aquifer porosity	13	13	
Counting uncertainty	25	30	
Well screen depth	50	50	
Representative concentrations of plume zone	35	35	
Radium background concentration	4	26	
TOTAL UNCERTAINTY (radium 226/228)	79	85	
TOTAL UNCERTAINTY (other contaminants)	79	81	

It can be seen that the estimated total uncertainty is approximately $\pm 80\%$ for all radionuclides, and both basins. The choice to use three-year maximum reported well concentrations, instead of average concentrations, was made in an attempt to bound the calculation uncertainty. Because the maximum results are 40-60% higher than the average results, and because the total uncertainty is estimated to be $\pm 80\%$, the use of maximum concentrations does not encompass the total concentration uncertainty. In addition, the total uncertainty estimated above does not include the additional problem of possible high bias in reported Ni-63 concentrations due to the presence of tritium.

3.2 Contaminants Sorbed to Formation

A full account of radionuclide activity in the saturated zone should consider the mass of sorbed contaminants on the surface of formation grains. In theory, calculation of sorbed activity is a simple matter, involving the soil/water partition coefficient (K_d) for each radionuclide. If the K_d for a specific constituent exceeds (formation porosity)/(bulk formation density), or 0.23 mL/g for FSB and HSB, then the activity of sorbed material will exceed the activity of dissolved material.

Soil/water partition coefficients are available for all radionuclides considered in this report, however, two issues makes their application at FSB/HSB problematic. First, most published K_ds

are intended for near-neutral pHs; however the most concentrated sections of the FSB/HSB plumes are quite acidic, with pHs as low as 3.2. Partition coefficients for radionuclides at these pHs are not widely available, and are probably much lower than corresponding K_{ds} for near-neutral solutions. If published near-neutral K_{ds} were used to estimate the inventory of sorbed contaminants, it is possible that an over-estimate of one or two orders of magnitude would result. Second, for some radionuclides at FSB, significant dissolved inventory resides in the swamp. An accurate assessment of sorbed activity would have to include partition coefficients between organic solids and water; few of these are available.

Therefore, the scope of this report is limited to calculation of the inventory of radiological contaminants which are dissolved in groundwater at FSB and HSB.

4.0 References

Carilli, J. T. and Golian, S. 2010. *Savannah River Site Composite Analysis Review Report*, Department of Energy Low-Level Waste Disposal Facility Federal Review Group Review Team, April 20, 2010.

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.

Killian, T.H., N.L. Kolb, P Corbo and I.W. Marine, 1985a, *Environmental Information Document F-Area Seepage Basins*, DPST-85-704, E.I. du Pont de Nemours and Company, Savannah River Laboratory, Aiken, S.C.

Killian, T.H., N.L. Kolb, P Corbo and I.W. Marine, 1985b, *Environmental Information Document H-Area Seepage Basins*, DPST-85-704, E.I. du Pont de Nemours and Company, Savannah River Laboratory, Aiken, S.C.

Looney, B.B., Eddy, C.A., Ramedeen, M., Pickett, J., Rogers, V., Scott, M.T. and Shirley, P.A., 1990, *Geochemical and Physical Properties of Soils and Shallow Sediments at the Savannah River Site (U)*, Westinghouse Savannah River Company, Aiken, S.C. 29808

Marcinowski 2010. Memorandum from Frank Marcinowski to Terrel Spears, July 16, 2010. *Approval of the Savannah River Site Department of Energy 435.1 Composite Analysis*, Rev. 0.

Phifer, M. A., Millings, M. R., and Flach, G. P. 2006. *Hydraulic Property Data Package for the E-Area and Z-Area Soils, Cementitious Materials, and Waste Zones*, WSRC-STI-2006-00198, Rev. 0, Savannah River National Laboratory, Washington Savannah River Company, Aiken, SC, September 2006.

SRNL 2010. Savannah River DOE 435.1 *Composite Analysis*, SRNL-STI-2009-00512, Revision 0, Volumes I and II, Savannah River National Laboratory, Aiken, SC. June 10, 2010.

SRNS 2015. Annual Corrective Action Report for the F-Area Hazardous Waste Management Facility, the H-Area Hazardous Waste Facility, and the Mixed Waste Management Facility (U). SRNS-RP-2015-00136, Volume I. Savannah River Nuclear Solutions, Aiken SC 29808, April 2015.

Wan J., T.K. Tokunaga, W. Dong, M.E. Denham, S.S. Hubbard, 2012. *Persistent Source Influences on the Trailing Edge of a Groundwater Plume, and Natural Attenuation Timeframes: The F-Area Savannah River Site.* Environmental Science and Technology 46, 4490-4497.

APPENDIX A

Groundwater Plume Maps for H-Area Seepage Basin





















This Page Intentionally Left Blank

APPENDIX B

Groundwater Plume Maps for F-Area Seepage Basin





















Distribution:

B. T. Butcher, 773-42A D. A. Crowley, 773-42A F. L. Fox, 704-59E N.V. Halverson, 773-42A T. Hang, 703-41A R. A. Hiergesell, 773-42A E.N. Hoffman, 999-W G. K. Humphries, 705-3C G. T. Jannik, 999-W K.M. Kostelnik, 773-42A W.P. Kubilius, 999-W C. M. Lewis, 730-4B M. G. Looper, 704-36E J. J. Mayer, 999-W M. R. Millings, 773-42A T. O. Oliver, 773-42A R. R. Seitz, 773-42A D. F. Sink, 704-56E H. M. Cardona (1 file copy & 1 electronic copy), 773-42A