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Contaminant Migration Modeling to Support the In Situ Decommissioning of Hardened Nuclear Facilities – 20215

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ABSTRACT

The decommissioning of hardened nuclear facilities provides a unique challenge in balancing current worker dose with exposure to future receptors and media. In situ decommissioning (ISD) is a cost-effective and safe option for the closure of such facilities but requires a detailed understanding of the threat that radioactive and hazardous constituents remaining in the facilities pose to groundwater and surrounding surface water. The Savannah River Site (SRS) extensively employs contaminant migration models within the decommissioning process to develop site-specific removal, grouting, and monitoring strategies in support of safe and effective final end states of nuclear facilities. This work discusses the use of contaminant migration modeling in the ISD process at SRS, including specific examples from the closure of P-Reactor and R-Reactor, Building 235-F, F-Canyon and F/H Laboratory Complex.

Migration models developed during the closure of P-Reactor and R-Reactor were divided into four source areas: the reactor vessel, the process area, the disassembly basin, and the purification wing. Each source area was assigned a specific inventory and migration pathway and was then subject to varying hypothetical removal, capping, and grouting schemes. Modeling indicated that groundwater Maximum Concentration Limits (MCLs) might be exceeded if no action was taken for eleven and ten constituents of concern (COCs) at P-Reactor and R-Reactor, respectively. These exceedances could occur in as few as 200 to 500 years. Alternatively, the migration modeling demonstrated that the selected ISD actions reduced contaminant mobility which allowed for significant radioactive decay and resulted in fewer predicted exceedances of groundwater MCLs (five COCs for P-Reactor; eight COCs for R-Reactor).

In response to the modeling results for P-Reactor and R-Reactor, effectiveness monitoring programs were developed to target contaminants, specific to each reactor, that may migrate to groundwater. Contaminant migration modeling also revealed that roof collapse was a large factor in the release of COCs to the environment, giving rise to roof improvements, and an inspection and vegetation control program to ensure roof stability over time.

Contaminant migration modeling is also aiding in the closure planning for hardened facilities in F Area. Building 235-F housed the Actinide Billet Line, which produced Np-237 billets for irradiation in SRS reactors, and the Plutonium Fuel Form (PuFF) facility that produced Pu-238 heat sources for the space program. As a result of these missions, areas within Building 235-F contain considerable residual amounts of both Pu-238 and Np-237. Contaminant migration modeling of Building 235-F was originally performed in 2012 to identify the feasibility of ISD and the amount of radioactive material removal required to prevent the exceedance of groundwater MCLs. The original model indicated that a 60% reduction in the PuFF facility Pu-238 inventory could keep groundwater concentrations below standards, while Np-237 did not pose a threat to groundwater. However, updates to the model with an emphasis on source impact pathways revealed that, due to the orientation of the source areas relative to groundwater flow, no amount of reasonable removal of Pu-238 would keep groundwater concentrations below MCLs and that Np-237 could be a large contributor to localized MCL exceedances. With this insight, the refined

2019 model is being used to assist in the development of grouting plans specific to each facility source area, where bentonite may be utilized to slow the migration of Pu-238 and its daughter products from the PuFF facility and a reducing grout may decrease Np-237 transport by ensuring the nuclide remains in the less mobile +IV oxidation state.

The beginning phases of contaminant migration modeling are underway for F-Canyon and associated facilities using lessons learned from the Reactors and Building 235-F. A contaminant migration pathway, similar to the pathway used for the reactor vessels, is being developed for the hot and warm canyons. The F-Canyon inventories are being spatially refined to identify specific and localized source zones, comparable to Building 235-F, that may impact groundwater.

Contaminant migration modeling provides vital input within the ISD process, from facility investigation to post closure effectiveness monitoring, making it a valuable tool in the development of safe and effective end states for hardened nuclear facilities at SRS.

INTRODUCTION

The Savanah River Site (SRS) encompasses 803 km^2 (310 mi^2) of South Carolina coastal plain uplands along the Savannah River in Aiken, Barnwell, and Allendale counties. The site was constructed during the early 1950s to produce the basic materials used in the fabrication of nuclear weapons, primarily tritium and Pu-239, in support of U.S. defense programs. The site included five reactors, which are more related to batch chemical reactors than commercial power generating reactors, and two separation areas that processed Pu-239 and tritium. Other processes, such as the fabrication of heat sources for space program applications, were also performed in the facilities in the separation areas. With the end of the cold war, a reduced requirement for defense related products resulted in the shutdown of the reactors and some of the chemical separation and fabrication facilities at SRS. These facilities, many of which are blast resistant hardened concrete structures, contain considerable amount of residual radioactive and hazardous material or hold-up, and must be closed in a manner that balances current worker dose with exposure to future receptors and media for years to come.

In situ decommissioning (ISD) is a cost-effective and safe alternative for the closure of hardened nuclear facilities. ISD can include the removal of source and the addition of barriers or cementitious material (grout) to minimize the release of residual material to the surrounding area as constituents slowly migrate to the water table. As such, ISD requires detailed migration modeling to understand the transport of residual radioactive and hazardous material over time. This work discusses the use of contaminant migration modeling at different times throughout the ISD process at SRS, including examples from the closure of P-Reactor and R-Reactor, Building 235-F, F-Canyon, and F/H Laboratory Complex.

DISCUSSION

Closure and long-term monitoring of P/R-Reactors

The nuclear reactors at SRS were built to manufacture Pu-239 and tritium on an industrial scale. P-Reactor began operations in February 1954. It was taken off-line for maintenance and safety upgrades in 1987, placed in warm standby in 1988, and placed in shutdown status in 1991. R-Reactor began operations in December 1953 and was shut down in 1964. Each reactor has been de-fueled, and all fissile materials have been removed. After shutdown, hold-up in each reactor building was primarily contained in three source areas: the Process Area including the Reactor Vessel, the Disassembly Basin, and the Purification Wing. R-Reactor also had residual source in the Emergency Basin. The Process Area is in the center of the reactor building and contains the reactor vessel, shield water system, control and safety rod actuating mechanisms, heat exchangers, primary coolant circuit pumps, helium blanket gas system, and the main control room. The Disassembly Area received assemblies after irradiation in the reactor. Irradiated fuel and target assemblies remained in the Disassembly Basin to allow the short-lived, high-activity isotopes to decay before chemical processing. To prevent the buildup of heat and fission products in the Disassembly Basin, the water was circulated through heat exchangers and a clarifier/filter to remove particulates. Tritium was also transferred to the Disassembly Basin water by moderator carry-over with the fuel and targets. Tritium buildup in the basin water was controlled by periodic purging to seepage basins. The Purification Area maintained moderator quality, where a portion of the moderator was continuously fed through area to remove light water, fission and activation products.

A number of alternatives for the closure of both reactors were investigated relative to cost, implementability, and effectiveness. The selected ISD was designed for each reactor to reduce contaminant mobility for a time period to allow significant radioactive decay [1&2]. Contaminant migration modeling revealed that roof collapse was a large factor in the release of hold-up to the environment, giving rise to roof improvements, and an inspection and vegetation control program to ensure roof stability over time. ISD for P-Reactor included: the removal of the Disassembly Basin above grade structure, grouting below grade portions of Disassembly Basin and construction of a sloped concrete cover over the grouted Disassembly Area; grouting all below grade spaces in the P-Reactor Building including Assembly, Process, and Purification Areas to grade; grouting the Purification Area cells; grouting the Reactor Vessel in place and constructing a concrete cover over the Reactor Vessel; removing the stack; and construction of modified roofs over select areas of the Process and Purification Areas, including the shield door slots, to prevent rainwater ingress [3]. ISD for P-Reactor is conceptualized in Figure 1.

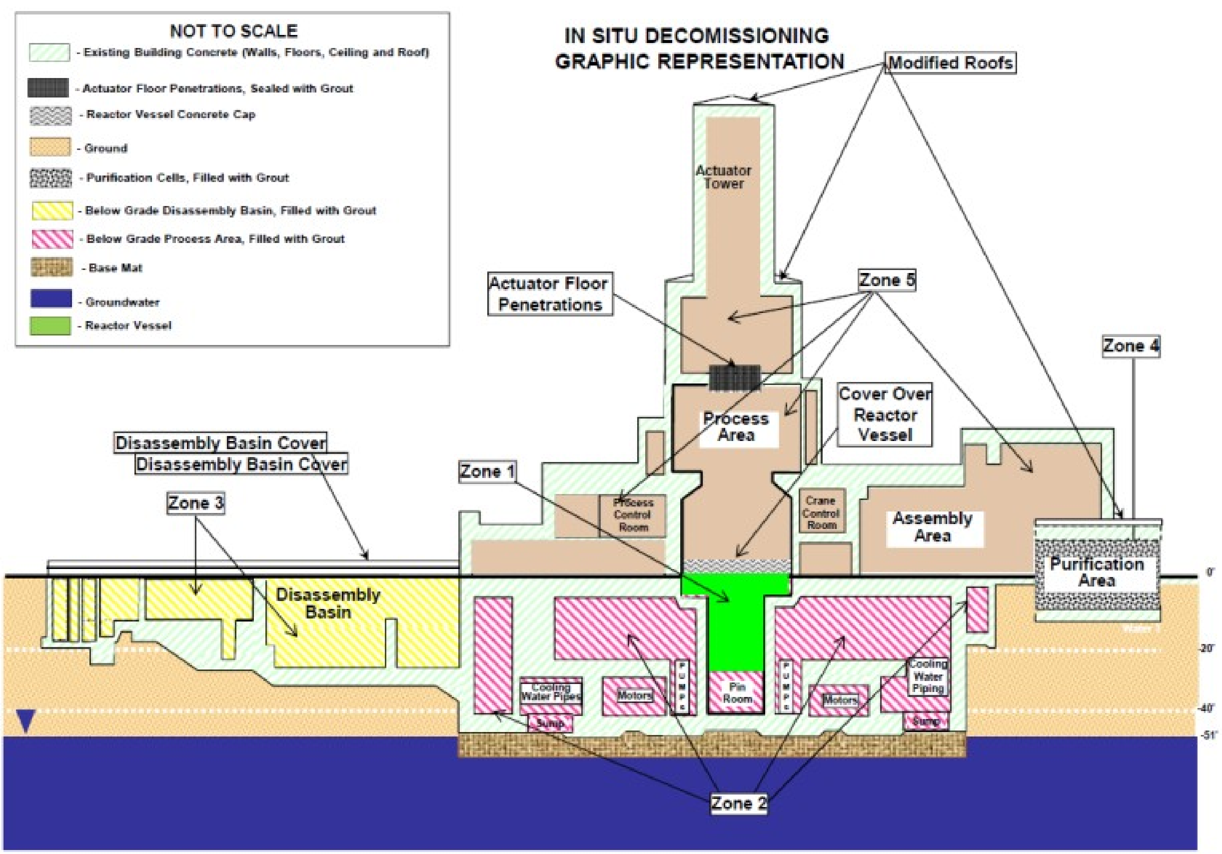

Figure 1. Representation of P-Reactor Building ISD

The selected ISD alternative for the R-Reactor included the following: grouting of below grade portions of the Reactor Building; grouting of the R-Reactor Vessel and placement of a concrete cover over the vessel; removal of the R-Reactor exhaust stack and consolidation of the stack debris in the Reactor Building prior to grouting; sealing of all exterior building openings, modifications of select roofs; grouting of the Disassembly Basin to grade; demolition of the above grade Disassembly Basin structure and placement of a concrete cover over the grouted Disassembly Basin and Area North of the Reactor Building [4].

Migration modeling in Goldsim (version 9.6) was performed for each ISD alternative as part of the selection process. A no action alternative where the Reactor Buildings were unaltered and abandoned was also included. P-Reactor Building complex was divided into four sources areas (disassembly basin, the purification wing, the reactor vessel, and the process area) [5], while R-Reactor had the same source areas with the addition of the emergency basin [6]. Each source was modeled in a separate sub-model. The expected receptor and point of assessment was groundwater 1 meter from the facility edge along a 1-dimensional cross-section of the source area. The cross section produced a maximum cumulative concentration for each subunit (Figure 2). A total of 86 constituents were considered which were mostly beta emitting activation products. Results indicated that even with the selected alternative exceedances of groundwater maximum concentration limits (MCLs) would occur. The exceedances were generally from mobile radionuclides within the Reactor Vessel and Disassembly Basin. The maximum concentration and peak time for the constituents of concern (COCs) predicted to exceed their respective MCLs are provided in Table 1. Exceedances for R-Reactor were largely similar to P-Reactor with the addition of I-129 (from the emergency basin), K-40, and elemental lead for the selected ISD alternative. Predicted maximum concentrations from R-Reactor were also lower due to the decreased operational time of R-Reactor.

These predicted exceedances gave rise to a targeted groundwater monitoring plan. The original monitoring program analyzed for all constituents that were predicted to exceed MCLs in the no action alternative to account for uncertainties in modeling [7&8]. This resulted in eleven analytes for P-Reactor, and ten for R-Reactor. Recently, the migration model was further used to refine the targeted radionuclides the most mobile COCs predicted to exceed MCLs under the no action alternative. C-14, Tc-99, and Cl-36 are analyzed for in groundwater near P-Reactor, while C-14, Cl-36 and I-129 are analyzed for near R-Reactor.

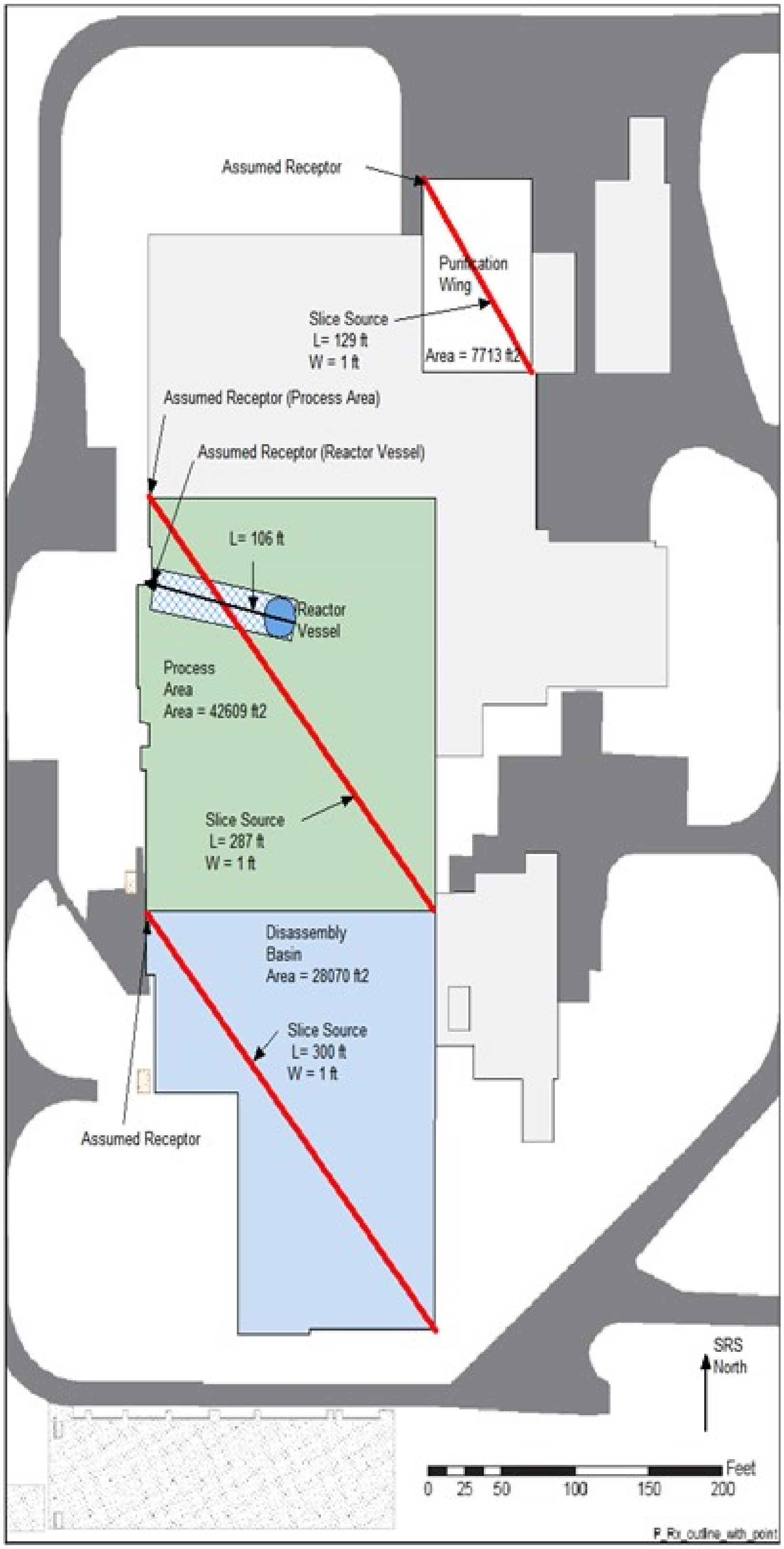

Figure 2. Modeled source areas within P-Reactor and locations of assumed receptors

Table 1. Maximum concentrations, peak times, and time of initial exceedance for COCs predicted exceed MCLs for P-Reactor.

Constituent	Concentration Time	MCL/PRG	Alternatives: No Action	Alternatives: ISD
C-14	Max. (pCi/L)	2,000	**220,000**	**6,000**
	Time of Peak (yr)		400	4,500
	Time of Initial Exceedance (yr)		220	1,800
Cl-36	Max. (pCi/L)	700	**21,000**	**1,200**
	Time of Peak (yr)		380	4,600
	Time of Initial Exceedance (yr)		220	1,800
K-40	Max. (pCi/L)	2	**3.9**	0.83
	Time of Peak (yr)		400	1,400
	Time of Initial Exceedance (yr)		300	--
Ca-41	Max. (pCi/L)	135	**230**	21
	Time of Peak (yr)		2,200	10,000
	Time of Initial Exceedance (yr)		300	--
Ni-59	Max. (pCi/L)	300	**28,000**	**6,400**
	Time of Peak (yr)		7,300	55,000
	Time of Initial Exceedance (yr)		520	25,000
Ni-63	Max. (pCi/L)	50	**1,900**	--
	Time of Peak (yr)		660	--
	Time of Initial Exceedance (yr)		420	--
Nb-93	Max. (pCi/L)	6	**510**	**28**
	Time of Peak (yr)		2,400	30,000
	Time of Initial Exceedance (yr)		280	2,000
Mo-93	Max. (pCi/L)	14	**3,300**	**64**
	Time of Peak (yr)		500	3,000
	Time of Initial Exceedance (yr)		280	2,000
Tc-99	Max. (pCi/L)	900	**1,100**	31
	Time of Peak (yr)		240	880
	Time of Initial Exceedance (yr)		220	--
Ag-108m	Max. (pCi/L)	6	**600**	0.032
	Time of Peak (yr)		1,400	4,400
	Time of Initial Exceedance (yr)		620	--
Total Dose	Max. (mrem/yr)	4	**560**	**85**
	Time of Peak (yr)		400	55,000
	Time of Initial Exceedance (yr)		220	1,300
Lead (stable)	Max. (pCi/L)	15	**17**	9.1
	Time of Peak (yr)		180,000	380,000
	Time of Initial Exceedance (yr)		90,000	--

Discussions towards the closure of Building 235-F

Building 235-F was constructed as part of the original Savannah River Plant in the early 1950s. The building is a windowless, reinforced-concrete structure with primary process areas located on the first floor and most support functions located on the second floor. Building 235-F houses the Plutonium Fuel Form Facility (PuFF), a metallography laboratory that supported the PuFF Facility, and the Plutonium Experimental Facility (PEF). These facilities were once used to fabricate heat sources from Pu-238 oxide powder for space program applications. Heat source fabrication processes were developed in PEF, and these processes were carried out on a large scale in the PuFF Facility. Another facility in Building 235-F, the Actinide Billet Line (ABL), was most recently used to fabricate billets from Np-237 oxide powder for irradiation in reactor targets. Extensive assays of 235-F have been performed that indicate significant radiological material (oxides of Pu-238 and Np-237) called "hold-up" remains within 235-F. The majority of the hold-up is located within PuFF and ABL. ISD alternatives are under consideration for decommissioning of Building 235-F in regard to groundwater protection, public/industrial worker protection, and cost. Discussions are currently ongoing with the regulators to select an ISD alternative for the final end state of Building 235-F.

Contaminant migration modeling of Building 235-F was originally performed in 2012 to identify the feasibility of ISD and the amount of radioactive material removal required to prevent the exceedance of groundwater MCLs. The model, developed in Goldsim (version 10.5) to provide stochastic results (1,000 realizations), investigated two ISD alternatives (grout first floor and grout the entire facility) and a no action alternative. The main driver for predicted exceedances was Ra-226, a daughter of Pu-238. The original model indicated that a 60% reduction in the PuFF facility Pu-238 inventory would keep groundwater concentrations below standards, while Np-237 did not pose a threat to groundwater.

In 2019, an update to the 2012 migration model for Building 235-F was performed. This update was performed deterministically in PORFLOW (version 6.42.9) and stochastically in Goldsim (version 12.5). An additional ISD alternative of grouting the whole facility with an installed sloped roof cap was included. Points of assessment included groundwater 1 meter from the facility, groundwater 100 meters from the facility, and surface water in a stream receiving groundwater from beneath the facility (683 meters from facility). The update placed emphasis on source term geometry, minimizing the source areas to the contaminated cells of each facility (Figure 3) and decreased the longitudinal dispersity by an order of magnitude to align with the current accepted values [9]. This refinement resulted in decreased dilution of the source and increased predicted concentrations in groundwater. The update also capitalized on the high level of hydrogeologic characterization for the region surrounding Building 235-F, which was previously incorporated into PORFLOW [10]. The deterministic run revealed that the extent of overlap for plumes emanating from the ABL and PuFF facilities is negligible. At points of assessment close to the facility, the plumes are spatially separated (Figure 4A) and at points further from the facility the chemical characteristics of the radionuclides result in temporal separation arising from relative difference in transport time between Pu-238 daughters and Np-237 (Figure 4B).

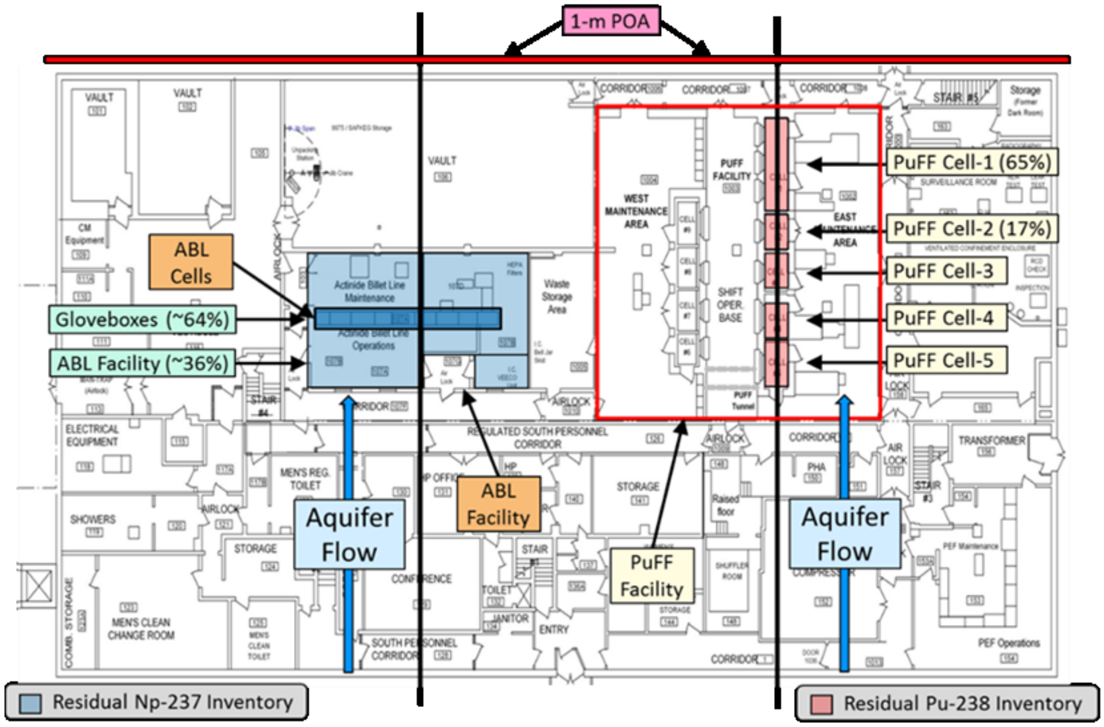

Figure 3. Location of source zones inside Building 235-F relative to aquifer flow and the 1-meter point of assessment. Percentages indicate the relative amount of inventory in each location (Pu-238 for PuFF and Np-237 for ABL).

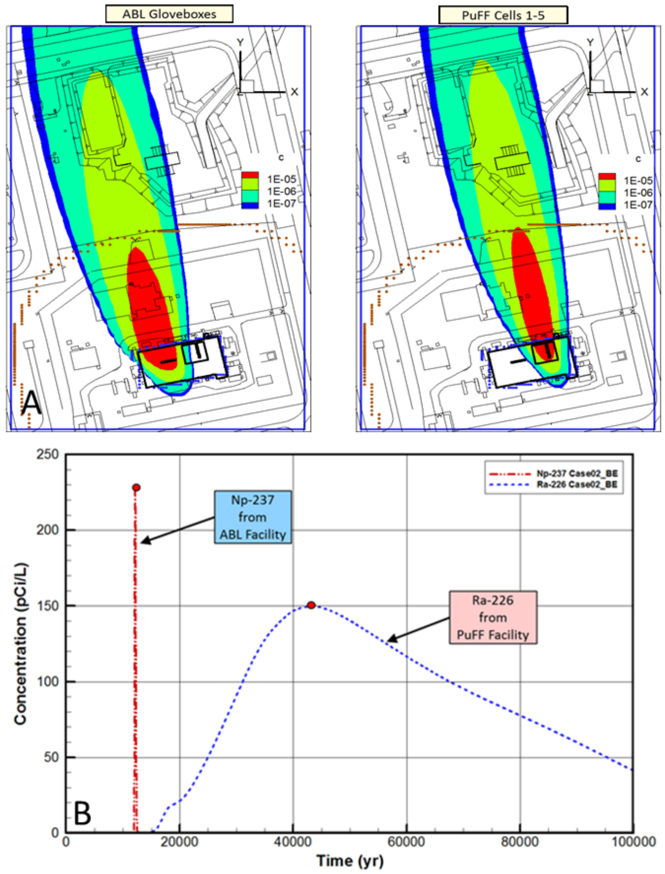

Figure 4. (A) PORFLOW-based tracer plume contours originating from the ABL and PuFF facilities. (B) PORFLOW-based predicted concentrations at the 100-m groundwater point of assessment illustrating the temporal plume separation between Np-237 and Ra-226 from ABL and PuFF, respectively.

The results from PORFLOW were then used to calibrate Goldsim, which has the capability to perform the necessary stochastic simulations (10,000 runs) to account for uncertainty in modeling long timeframes. Overall, the model results predicted that over a 100,000-year timeframe MCLs would be exceeded in groundwater but little impact to surface water was expected. The exceedances generally occurred after 10,000 years. The main exceedance drivers were U-234 and Ra-226, both daughters of Pu-238, for the PuFF facility, and Np-237 for ABL. These radionuclides result in exceedances of the gross alpha (Table 2), radium, and beta-gamma dose MCLs for the plume emanated from the PuFF facility, while the plume emanating from ABL exceeds the gross alpha (Table 2) and beta-gamma dose MCLs. The model also established that predicted concentrations are linear with residual inventory, allowing for the calculation of a percent removal required for each facility (ABL and PuFF separately due to peak separation) to maintain concentrations below MCLs (Table 2). The calculated percent removals indicate that extensive material removal would be required to maintain groundwater concentrations below MCLs. These results are currently being used in discussions with the regulators to select an ISD alternative that balances current worker dose with exposure to future receptors and media.

Table 2. Peak concentrations, time to peak, and percent removal required to maintain concentrations below the gross alpha MCL for three ISD alternatives and a no action alternative for both the PuFF (Pu-238 daughters) and ABL (Np-237) facilities. Exceedances are highlighted.

		PuFF Gross Alpha MCL = 15 pCi/L			ABL Gross Alpha MCL = 15 pCi/L		
ISD Alternative	POA (m)	Conc. pCi/L	Time to Peak(yrs)	Percent Removal to meet MCL	Conc. pCi/L	Time to Peak(yrs)	Percent Removal to meet MCL
No action	1	3259	25260	99.5%	542	1340	97.2%
	100	363	30860	95.9%	109	1410	86.2%
	Surface	0	53350	0.0%	0	1510	0.0%
Grout first floor	1	3579	32960	99.6%	151	5720	90.1%
	100	389	38450	96.1%	31	5790	51.5%
	Surface	0	60660	0.0%	0	5890	0.0%
Grout entire 235-F	1	3965	42060	99.6%	41	12740	63.0%
	100	418	47060	96.4%	8	12800	0.0%
	Surface	0	68460	0.0%	0	12890	0.0%
Grout entire 235-F with cap	1	4150	72260	99.6%	23	21760	33.7%
	100	415	76960	96.4%	5	21830	0.0%
	Surface	0	97760	0.0%	0	21930	0.0%

Informing the characterization of F-Canyon and F/H Laboratory Complex

F-Canyon was constructed in the early 1950s as part of the original Savannah River Project and was used as one of two radiochemical processing facilities at SRS. The Plutonium-Uranium Extraction process was used in F-Canyon to separate and recover Pu-239 and U-238 from irradiated materials, and to stabilize plutonium residues inside two main cells known as the “hot” and “warm” canyons. F-Canyon was shut down in 1991 after the Cold War ended; though operations began briefly again in 1996 to stabilize nuclear materials until 2002 when the last production run was completed. The F/H Laboratory Complex (also known as the Central Laboratory Facility) consists of Buildings 772-F, 772-1F, and 772-4F which began operations in 1954, 1987, and 1993, respectively. The primary mission of the F/H Laboratory Complex for the last 45 years has been to support the chemical separations processing activities of F-Canyon and H-Canyon via chemical and radiological testing of material samples. The laboratories are currently being deactivated.

ISD alternatives are under consideration for decommissioning of the F-Canyon and F/H Laboratory Complex in regard to groundwater protection, public/industrial worker protection, and cost. To evaluate ISD alternatives, the development of contaminant fate and transport models for each facility is required. However, a limited understanding of the current radiological hold-up inventories is known for each of the facilities. With the recent attention on the source term locations in the modeling of Building 235-F, the facility footprint and migration paths must be handled carefully for meaningful and defendable results to be obtained from migration modeling. For instance, the current F-Canyon hold-up is based on the lifecycle process inventory difference and assumed to be dispersed throughout the facility, but groundwater modeling would be quite sensitive to this assumption. The presence of localized hot spots (i.e., sub-sections of F-Canyon having an associated high inventory) can increase the concentration at the water table when compared to modeling a uniformly distributed source term. Similarly, the deactivation of F/H Laboratory Complex is resulting in a decrease in residual radiological hold-up, which must be characterized to an extent that leads to accurate groundwater modeling results. As such, a contaminant migration model is currently being developed to understand the spatial definition needed in both the F-Canyon and F/H Laboratory Complex to provide accurate and defensible migration modeling results. The model will stochastically vary the quantity and location of the inventory for both facilities to establish best- and worst-case scenarios for inventory locations and establish whether further characterization is required.

CONCLUSIONS

From characterization and deactivation planning, closure discussions, and effectiveness monitoring, migration modeling provides vital information throughout the ISD process for hardened nuclear facilities. Accurate models that stochastically account for uncertainties can predict the release of residual hold-up in facilities that are closed through ISD. These predictions provide important information so that a protective and cost effective ISD alternative is selected to support the safe and effective final end states of nuclear facilities.

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