Fate and Transport of Contaminants at Soils Sites - 15512

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ABSTRACT

As part of the Soils Activity of the Environmental Management Operations at the NNSA, Nevada Field Office, radionuclide-contaminated Soils sites will undergo regulatory closure in accordance with the 1996 Nevada Federal Facilities Agreement and Consent Order. One closure alternative that has been evaluated for each Soils site has been "closure-in-place" with fencing and posting, along with administrative controls. However, potential disturbance agents for radionuclide-contaminated soils left in place are possible erosion and transport by wind and water. Previous work with radionuclide-contaminated soils has indicated that the highest activity levels of Pu tend to be found in the fine sands, silts, or clays, which are the most likely particle sizes to be transported by wind re-suspension, and by runoff flow events. Monitoring and sampling stations have been deployed at specific Soils sites to collect meteorological data, airborne particulate filter samples, and both suspended and bedload sediment transport samples during and after runoff flow events, as needed. The primary objective of the air monitoring and sampling stations is to evaluate whether there is wind transport of radionuclide-contaminated soils from Soils sites. The primary objective of the precipitation and runoff flow data collection, as well as both suspended and bedload sediment sampling, is to determine during which rainfall events (*i.e.*, precipitation return interval, duration, and intensity) radionuclide-contaminated soils are likely to be transported. The NNSS continues to demonstrate compliance with US DOE Order 458.1 for the Radiation Protection of the Public and the Environment, as reported in annual environmental reports [1].

INTRODUCTION

In the late 1950s and early 1960s, the US DOE (formerly the Atomic Energy Commission [AEC]), conducted a series of nuclear device safety tests or weapons effects tests, sometimes known as Plutonium (Pu) dispersal tests, on the NNSS (formerly the Nevada Test Site), and on the Tonopah Test Range (TTR) and an adjacent area of the Nevada Test and Training Range (NTTR) (formerly the Nellis Air Force Range) to the north of the NNSS. These tests were conducted using conventional explosives detonated in the presence of nuclear devices to assess the dispersal of radionuclides, to evaluate the effectiveness of storage structures to contain ejected radionuclides, and to assess potential decontamination techniques.

On the NNSS, safety tests were conducted at numerous locations, including at Plutonium Valley and at the Smoky Site (Figure 1). Plutonium Valley, located in Area 11 of the NNSS in the southeastern part of Yucca Flat, Nevada, just east of Yucca Lake (playa lake), was the site of the Project 56 tests in 1955 and 1956. Project 56 consisted of a series of four nuclear device safety



Figure 1. Environmental monitoring instrumentation is installed at both Plutonium Valley and the Smoky Site, two locations where safety tests were conducted on the NNSS.

tests detonated at four separate test beds within the valley. The tests left alpha contamination scattered over the ground surface near the test beds, and subsequent ground and aerial surveys [2 and 3] indicate that radionuclide-contaminated soils have migrated by surface water transport from some of the test beds. In 1958, three safety tests - Oberon, Ceres, and Titania – were conducted at the Smoky Site (named for an above ground nuclear device test detonated in 1957), followed in 1964 by a weapons effect test, Mudpack. The tests left alpha contamination scattered over the ground surface. An aerial survey [3], as well as anecdotal information [4], suggests that radionuclide-contaminated soils have been migrating down ephemeral channels that traverse the Smoky Site, and in some cases, may have been deposited on the road bordering the southeast boundary of the Smoky Site. Operation Roller Coaster, conducted in 1963 on the TTR and adjacent NTTR land, consisted of four tests. Three tests – Clean Slates I, II, and II – were conducted on the TTR in Cactus Flats and resulted in dispersal of Pu over the ground surface downwind of the ground zero location of each test (Figure 2). The fourth test, conducted on adjacent NTTR land, is not part of this study.

DESCRIPTION

As part of the Environmental Management Operations Soils Activity, the US DOE, NNSA, Nevada Field Office (NNSA/NFO), requested that the Desert Research Institute (DRI) construct, deploy, and maintain environmental monitoring stations (inclusive of meteorological and soils conditions) at the Plutonium Valley, Smoky Site, and TTR Soils sites. These environmental monitoring stations are inclusive of meteorological and soil condition measurements, including temperature, precipitation, barometric pressure, relative humidity, solar radiation, wind speed and direction, etc., as well as soil temperature and moisture content (Figure 3). The objectives of the monitoring efforts are to evaluate whether there is wind or water transport of radionuclidecontaminated soils from the Soils sites, and if so, under what environmental conditions does the transport occur.

In addition to the environmental monitoring at each site, site-specific monitoring equipment was also installed at each studied Soils site to address site-specific contaminant migration concerns. At Plutonium Valley, to address the migration of radionuclide-contaminated soils in the channel emanating from the southernmost test location, an Isco sampler (Teledyne Isco, 4700 Superior Street, Lincoln, NE) was installed to collect suspended-load samples (fine-sized soil particles entrained within the water column) during flow events. Bedload traps were installed in the channel bottom to collect larger sized soil particles that are transported by saltation along the bed during flow events (Figure 4). Both an ultrasonic depth sensor and pressure transducer were installed to determine flow depths through measured channel cross-sections, which then can be used to estimate runoff discharge volumes through the channel reach. In addition, real time air particulate samplers (Figure 3) were installed to determine size fractions of dust particles emitted from the sites; however, no filter-based air monitoring is performed in Plutonium Valley.

Transport of radionuclide-contaminated soils during runoff-response events is also a recognized problem at the Smoky Site. To address this concern, a flume was installed in a channel emanating from the contamination area, to estimate runoff discharge in response to measured precipitation events (Figure 5). To determine if radionuclide-contaminated soils are being transported by flows, bedload grab samples are collected after runoff events, at various locations



Figure 2. The TTR environmental monitoring stations are located on the south side of the Sandia National Laboratory compound (located adjacent to the airport), and at the north ends of the Clean Slate I and III contamination areas.



Figure 3. An example of a typical environmental monitoring station (tower structure shown on the trailer), inclusive of meteorological and soil conditions monitoring equipment. Environmental monitoring station equipment not shown in this figure includes solar panels and batteries, and a buried soil moisture probe. A particle profiler, low-volume air sampler, and pressurized ion chamber (PIC) are also installed on this station, with the first two items used for air monitoring, and the third for radiological monitoring.

within the channel and overbank areas, although no suspended-load sampling is performed at this location. No air monitoring or particulate sampling is performed at the Smoky Site location.

At the TTR sites, air monitoring stations were installed to evaluate whether there is wind transport of radionuclide-contaminants, specifically Pu, from the Operation Roller Coaster sites; and if so, under what meteorological and soil conditions does such transport occur. The fundamental design of these stations is similar to that used in the Community Environmental Monitoring Program (CEMP) stations surrounding the NNSS; thus, both real time air particulate samplers and air monitoring (*i.e.*, filter samples) equipment was installed (Figure 3). The real time air samples monitor for fine particulate matter ($0.5 \mu m$ to $10 \mu m$ in diameter) suspended in the air. Saltation sensors, which monitor for wind-induced movement of sand-sized particles (>100 µm), are also installed at each station. In addition to the air monitoring stations designed primarily to detect Pu, pressurized ionization chambers (PICs) were installed to detect the gamma energy emitted during decay of Americium (Am)-241, a daughter product of Pu-241. Thus, in addition to the routine environmental monitoring equipment, two radiological data collection systems are deployed at each of the TTR monitoring stations. At the present time, no surface water runoff monitoring is performed at the TTR sites.

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Figure 4. In Plutonium Valley, the Isco sampler and ultrasonic depth sensor are installed in the channel emanating from the southernmost test bed (top left). A close-up of the Isco sampler, which collects suspended-load samples during flow events, is shown (top right). A bedload sampler is located downstream of the ultrasonic depth sensor (top right corner of bottom left photo), with a close-up of the bedload sampler shown (bottom right).



Figure 5. View looking downstream at the flume installed in a channel on the Smoky Site. A pressure transducer (used to measure flow depth) is installed at the flume, and the data are transferred to an environmental monitoring station located just off of the right edge of the photo.

DISCUSSION

The primary objective of the precipitation and runoff flow data collection, as well as both suspended and bedload sediment sampling, is to determine during which rainfall events (*i.e.*, precipitation return interval, duration, and intensity) radionuclide-contaminated soils are likely to be transported, and by which mechanism, and in what concentrations. Flyover surveys, using Am-241 as an indicator for Pu particulate transport, revealed that some surface transport of radionuclide-contaminated soils has already occurred along channels (Figure 6) near two of the test beds in Plutonium Valley, as well as channels emanating from the Smoky Site, suggesting additional surface movement of radionuclide-contaminated soils is possible in the future at these locations. Clay and silt particles associated with Pu contamination can be easily entrained within channelized flows as suspended sediment, and may be transported significant distances. In contrast, fine sand particles can be transported as bedload, and thus may remain more localized to the contamination source. Assuming a "closure-in-place" alternative is implemented, transport of radionuclide-contaminated soil by runoff flow events may necessitate changes to both physical and administrative closure boundaries. Surface water modeling results provide rationale for characterization of specific Soils sites with respect to transport of radionuclide-

contaminated soils by water as part of implementation of the site-specific Corrective Action Investigation Plan.

Measurable precipitation events, along with any resulting ephemeral runoff events, are infrequent in semi- and arid environments; thus, there are long periods where no runoff and sediment transport measurements are collected. However, monsoonal precipitation events during the summer of 2013 resulted in several measurable runoff events at the Smoky Site where runoff flow data and sediment samples were collected.



Figure 6. (*Left*) Am-241 detections in Plutonium Valley suggest migration of radionuclidecontaminated soils along channels conveying runoff away from the Soils sites. Items numbered \ 1 through 4 identify the four Project 56 ground zero sites; items 5 and 6 identify plumes in channels draining the ground zero areas; and item 7 designates an isolated low activity spot (after [3]). (*Right*) Am-241 detections at the Smoky Site also suggest migration of radionuclidecontaminated soils along channels away from the Soils sites [3], as evidenced by the lobe of measurable contaminated material extending southeastward from the contaminated area associated with the Mudpack weapons effect test (marked as number 3). Numbered items 1, 2, and 4 identify ground zero for three safety experiments: Oberon, Ceres, and Titania, respectively.

During storms on both 24 and 28 July 2013, runoff was recorded through the Smoky Site flume and sediment was subsequently deposited downstream of the flume. At the Smoky Site environmental monitoring station precipitation gage, 1.01 and 1.38 inches of total (24-hour) precipitation was recorded on 24 and 28 July, respectively. The duration of the first storm was subsequently determined to be less than 2 hours, with a maximum one-hour precipitation intensity of 0.87 inches/hour. The second storm slightly exceeded 4 hours in duration, with a maximum intensity of 0.84 inches/hour. When referenced to the National Oceanic and Atmospheric Administration (NOAA) Atlas 14, Precipitation Frequency Data Server (PFDS) [5] (<u>http://hdsc.nws.noaa.gov/hdsc/pfds/pfds_map_cont.html?bkmrk=nv</u>; accessed 6 September 2013), both storm maximum intensities approximate a 10-year, 1-hour (60-minute) event. In terms of precipitation frequency, again referencing the NOAA Atlas 14 PFDS [5], the 24 July storm approximated a 10-year, 2-hour storm; whereas the 28 July storm approximated a 10-year, 6 hour storm [6]. (Clark County Regional Flood Control District [7], which provides the closest hydrologic engineering design guidance available to the NNSS, uses 6-hour design storms.) Neither storm exceeded a 2-year, 24-hour point precipitation depth-duration-frequency value [5].

Subsequent to these late summer 2013 storms at the Smoky Site, sediment samples were collected from locations downstream of the flume and sent to appropriate soils laboratories licensed to handle radiological material to determine particle size distribution, as well as analyze for radionuclides, and provide correlation of any such contamination with particle sizes [6]. For the isotope analysis, each sediment sample was subdivided into three separate size classes consisting of material less than 63 μ m; material equal or greater than 63 μ m, ranging up to 250 μ m; and material greater than 250 μ m (Table I). The two smaller size classes were then analyzed to establish the associated concentrations of Am-241, Pu-238, and Pu-239/240 (Table I). (The largest size class, consisting of material greater than 250 μ m was not analyzed for radionuclide concentration.) In almost all cases, the smallest particle size class (<63 μ m) was associated with the highest radionuclide concentrations.

	Particle Size Class Percentages			<63 µm (pCi/g)			63 μm to 250 μm (pCi/g)		
Sample	<63 µm (%)	63 to 250 μm (%)	>250 µm (%)	Am- 241	Pu- 238	Pu- 239/240	Am- 241	Pu- 238	Pu- 239/240
SMK01	3.8	12.0	84.2	1.02	0.21	5.87	0.35	0.23	2.14
SMK02	1.9	6.6	91.5	1.96	0.28	9.87	0.33	0.11	1.20
SMK03	1.4	1.6	97.0	1.48	0.25	13.30	0.70	0.09	1.42
SMK04	17.4	23.9	58.7	1.07	0.53	24.80	0.59	0.24	3.65
SMK05	1.6	6.3	92.1	0.92	0.37	7.00	0.28	0.28	1.41
SMK 06	6.7	11.9	81.4	2.29	0.37	8.37	0.39	0.09	1.84
SMK07	1.5	1.5	97.0	0.84	0.23	4.03	0.99	0.22	5.75
SMK08	6.5	18.4	75.1	1.67	0.24	7.40	0.46	0.13	1.83

Table I. Particle size class percentages and associated radionuclide concentrations for sediment samples collected subsequent to the 2013 rainfall-runoff events at the Smoky Site [6].

Three Analysis of Variance (ANOVA) statistical computations were conducted to determine whether any of the specific radionuclides were binding to the soil preferentially between the two smaller particle size classes (Table II). The null hypothesis (*i.e.*, the premise is true) for the analysis was that the mean concentration values for each radionuclide associated with the different soil particle size classes were statistically the same. The alternative hypothesis (*i.e.*, the premise is false) was that the mean concentration values for each radionuclide associated with the different particle size classes were statistically different. An F-ratio comparing the concentration of each radionuclide between particle size classes was calculated, and compared against the tabled F_{crit} value [8]. In all cases, the calculated F-ratio value exceeded the tabled F_{crit} value; therefore, the mean concentration values differed (*i.e.*, the null hypothesis was determined to be false), which quantitatively demonstrates that the radionuclides preferentially bind to the finer particles. Similar findings were reported for bedload samples collected at Plutonium Valley.

Preferential binding of Pu and Am isotopes to the smaller sized soil particles [2] has major implications concerning movement of the isotopes in the environment. The smaller particles are more mobile during runoff events in response to even relatively frequent, but non-substantial, rainfall events, such as the 24 and 28 July 2013 storms with 2- or 10-year return intervals (depending on duration). Less energy is required for movement so smaller sized soil particles are more likely to enter the flow stream (wash load). The energy associated with the velocity of the flows will not only start the particles moving, but it will keep the particles in suspension once entrained in the flow. Flow velocities can fluctuate greatly, and the larger particle sizes will fall out of suspension at lower velocities, and will become part of the bedload. Data on suspended and bedload sediment transport will aid in assessing the likelihood that physical and administrative closure boundaries may be impacted by surface water flows at specific Soils sites where the "closure-in-place" alternative is used. The fine particle sizes are also susceptible to movement via wind; therefore, aeolian transport of the smaller particles represents a greater risk of radionuclide-contaminated soil movement.

Inhalation of airborne Pu particles is the exposure path most likely to result in health effects in humans. Because Pu particles tend to attach to smaller sized soil particles, suspension of contaminated dust is the most likely mechanism for human exposure and also the most likely mechanism for transport beyond the known extent of contamination. Suspension and transport

Table II. Mean and standard deviation of each radionuclide concentration (pCi/g) per particle size class for sediment samples from the Smoky Site. Using an Analyis of Varienace (ANOVA) analysis, in all cases, the calculated F-ratio value exceeded the tabled F_{crit} value; therefore, the mean values differed, which quantifies that the radionuclides preferentially bind to the finer particles [6].

	<63 µm		63 to 250 µm		F-ratio	
Radionuclide	μ	σ	μ	σ	Tabled F _{crit}	Calculated F
Am-241	1.4	0.5	0.5	0.2	3.1	18.9
Pu-238	0.3	0.1	0.2	0.1	3.1	8.6
Pu-239/240	10.1	6.6	2.4	1.6	3.1	10.4

of contaminated dust, as well as near-bed movement of fine sands by creep-reptation and saltation processes, are influenced by local meteorological and other environmental conditions, such as wind speed and soil moisture content. The primary objective of the air monitoring and sampling stations is to evaluate whether there is wind transport of radionuclide-contaminated soils from Soils sites, and if so, under what conditions it occurs.

From the TTR environmental monitoring and air sampling stations, wind and suspended dust particle data analyses show the average concentration of respirable-size soil particles (*i.e.*, particulate matter of aerodynamic radius of less than 10 micro-meters, or PM₁₀) increases in an approximately exponential pattern with linear increases in wind speed (Figure 7). A significant increase in PM₁₀ concentration begins at approximately 40 km/hr (25 mph, hourly average). Relatively small increases in wind speed above this threshold may result in significant increases in PM₁₀ concentration. However, even though PM₁₀ concentration increases dramatically at high wind speeds, this does not imply that significantly large volumes of soil material are moving from contamination areas. Several factors were determined to limit suspended dust volumes. High wind speeds at or above 40 km/hr (25 mph) occurred less than five percent of the time at the monitoring and sampling stations, and although high wind speeds may continue for a sustained period, PM₁₀ concentration tended to drop off over time, as the PM₁₀ supply is source limited (e.g., all available dust is transported during the early part of a wind event and little additional dust is available until other phenomena again break up the soil surface). In addition, because wind events are regional in scale, large areas upwind of the monitoring stations, including but not limited to the contamination areas, are capable of contributing to the PM_{10} concentration measured at the air monitoring stations. Thus, the PM_{10} measured at an air monitoring station may have no or little contribution from the contamination area.



Figure 7. The PM_{10} trends as a function of wind speed for TTR stations 400, 401, and 402 for calendar year 2013 [10].

Airborne dust particles were collected continuously at each of the TTR air monitoring stations as the air samplers draw ambient air through filters so the dust gathers on the filters. The filters are exchanged every two weeks and sent to the laboratory for gross alpha, gross beta, and gamma spectroscopy analyses. The gross alpha, gross beta, and gamma spectroscopy analyses results for calendar year 2013 are presented in Tables III, IV, and V, respectively.

Sampling Location	Number of	Concentration (x10 ⁻¹⁵ μ Ci/mL [3.7 x 10 ⁻⁵ Becquerel (Bq)/m ³])				
	samples	Mean	Standard Deviation	Minimum	Maximum	
Station 400(G)	26	2.03	1.01	0.66	5.69	
Station 400(C)	15	1.33	0.64	0.18	2.63	
Station 401(G)	19	1.60	0.67	0.55	3.08	
Station 401(C)	6	1.05	0.30	0.70	1.63	
Station 402(G)	19	2.04	0.80	0.35	3.65	
Station 402(C)	7	1.70	0.61	0.89	2.96	

Table III.Gross alpha results for TTR sampling stations for calendar year 2013 [10].

NOTES: Bq = Becquerel; m^3 = cubic meter; μ Ci/ml = microcurie per milliliter; TTR = Tonopah Test Range; (G) = glass filter; (C) = cellulose filter; glass-fiber filters retain particulates greater than 0.3 µm; cellulose-fiber filters retain particulates greater than 20 µm.

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Sampling Location	Number of	Concentration (x10 ⁻¹⁴ µCi/mL [3.7 x 10 ⁻⁴ Becquerel (Bq)/m ³])					
	samples	Mean	Standard Deviation	Minimum	Maximum		
Station 400(G)	26	2.03	0.44	1.26	3.25		
Station 400(C)	15	1.12	0.22	0.73	1.54		
Station 401(G)	19	1.64	0.44	0.46	2.30		
Station 401(C)	6	0.82	0.25	0.55	1.24		
Station 402(G)	19	2.13	0.67	0.68	3.34		
Station 402(C)	7	1.05	0.29	0.69	1.53		

NOTES: Bq = Becquerel; m^3 = cubic meter; μ Ci/ml = microcurie per milliliter; TTR = Tonopah Test Range; (G) = glass filter; (C) = cellulose filter; glass-fiber filters retain particulates greater than 0.3 µm; cellulose-fiber filters retain particulates greater than 20 µm.

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ION	Station 400	Station 401	Station 402
Beryllium-7 (Be-7)	26	18	26
Lead-210 (Pb-210)	21	9	16
Potassium-40 (K-40)	7	7	3
Protactinium-234m (Pa-234m)	1	0	0

Table V. The number of CY2013 particulate samples in which naturally occurring radionuclides were identified by gamma spectroscopy varied by radionuclide and between the TTR sampling stations [10].

Gross alpha and gross beta concentrations reported for CY2012 for CEMP stations [10] surrounding the TTR are used for comparison. The mean gross alpha values for the TTR stations were equivalent to or about 27 percent higher than the highest value observed at the surrounding CEMP stations. The mean gross beta value for Station 402 was approximately equivalent to the highest value reported for a CEMP station, located at Sarcobatus Flats. The mean gross beta values for Station 400 were slightly below this level and the value for Station 401 was near the low end of the values reported for the surrounding CEMP stations.

Gamma spectroscopy identified only naturally occurring radionuclides in the particulate samples collected from TTR air monitoring Stations 400, 401, and 402 during CY2013 (Table V), as well as during CY2012 at the surrounding CEMP stations [10]. The detected radionuclides occurred with varying frequency, with Be-7 and Pb-210 being the most commonly detected. No anthropogenic, gamma-emitting radionuclides were detected. No indicators of Pu were detected. The similarity between TTR and CEMP radiological observations suggests that there is no transport from the Clean Slate sites, that radiation at the TTR monitoring stations is due to natural (terrestrial and cosmic) sources, and that the levels of radiation observed are approximately equivalent to levels observed at the surrounding CEMP stations.

Gamma radiation is measured using a PIC detector. The PIC detectors are generally deployed to detect gamma radiation events that substantially exceed ambient radiation levels as a result of human activities. In the absence of such activities, ambient gamma radiation rates are reported. These radiation values vary naturally among locations, reflecting differences in altitude (cosmic radiation) and radioactivity in the soil (terrestrial radiation). Additionally, slight variations in gamma radiation at a single location may be due to changes in weather [11].

Intervals of increased gamma values were associated with meteorological conditions that indicate passage of storm fronts and typically include precipitation events [10]. The high dust counts observed prior to the intervals of increased gamma values are likely due to the increased wind speed associated with the storm front passage. Winds associated with the intervals of increased gamma values were variable, changing direction, or from the northwest. The difference between the high gamma values and the preceding observations were similar despite the differences in wind direction. As the TTR Stations 401 and 402 are located at the north end of the areas of contamination, winds from the northwest would transport uncontaminated soils rather than soils from the contamination areas as they reached the monitoring stations. Therefore, it appears the observed intervals of higher gamma values are not due to resuspension and transport of soil particles from the contamination areas.

Although the site-specific wind and water erosion and transport investigations are conducted as part of the Nevada Federal Facilities Agreement and Consent Order process, the US DOE also demonstrates compliance with applicable radiation regulations, notably US DOE Order 458.1 (Radiation Protection of the Public and Environment) and the Clean Air Act's National Emission Standards for Hazardous Air Pollutants (NESHAP). Monitoring of air, water, direct radiation, and biota from locations across the NNSS consistently leads to an estimated total dose to a maximally exposed member of the public of a small fraction of the dose limit [1]. At the TTR, past monitoring demonstrates values below the NESHAP threshold that requires continuous air monitoring [12].

CONCLUSIONS

As "closure-in-place" is a commonly used regulatory closure practice at many of the NNSS Soils sites, understanding the potential disturbance agents for radionuclide-contaminated soils left in place is important to ensure that physical and administrative boundaries are not compromised. Previous work [2] with radionuclide-contaminated soils has indicated that the highest activity levels of Pu tend to be found in the fine sands, silts, or clays, which are the most likely particle sizes to be transported by wind re-suspension or saltation, and by runoff flow events. Monitoring and sampling stations deployed at specific Soils sites to collect meteorological data, airborne particulate filter samples, and both suspended and bedload sediment transport samples during and after runoff flow events, as needed, have demonstrated that contaminant migration by wind transport is limited; however, at some sites where ephemeral channels intersect the contamination areas, radionuclide-contaminated soils migration has occurred along the channels during flow events. Thus, precipitation event-based monitoring can be prescribed as an effective contaminant transport monitoring strategy at these specific sites. Assuming a "closure-in-place" alternative is implemented, transport of radionuclide-contaminated soil by wind and water erosion and transport processes may necessitate changes to both physical and administrative closure boundaries.

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