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MEASUREMENTS OF PLUTONIUM AND AMERICIUM IN SOIL SAMPLES FROM PROJECT 57 USING THE SUSPENDED SOIL PARTICLE SIZING SYSTEM (SSPSS)

prepared by
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EXECUTIVE SUMMARY

As part of the preliminary site characterization at Project 57, soil samples were collected for separation into several size-fractions using the Suspended Soil Particle Sizing System (SSPSS), designed and fabricated by the Division of Atmospheric Sciences (DAS) of the Desert Research Institute (DRI). SSPSS operations, consisting of particle separation by sieving and aerodynamic sizing, were performed by personnel of the Harry Reid Center for Environmental Studies (HRC) at the University of Nevada, Las Vegas (UNLV).

Soil samples were collected specifically for separation by the SSPSS at three general locations in the deposited Project 57 plume, the projected radioactivity of which ranged from 100 to 600 pCi/g. The primary purpose in focusing on samples with this level of activity is that it would represent anticipated residual soil contamination remaining at the site after corrective actions are completed. Consequently, the results of the SSPSS analysis can contribute to dose calculation and corrective action-level determinations for future land-use scenarios at the site. Higher activity soils were also not considered because of restrictions on the amount of radioactive material that UNLV is licensed to have in its possession. Soil samples were collected at five sites for each of the three locations. At each sample site, vegetation and debris were removed and soil was collected to a depth of approximately 8 cm, placed in a 2-liter container, and marked with a sample identification number. At two of the sample sites, a second soil sample was collected to serve as a duplicate sample. A total of 17 samples were collected for particle size analysis using the SSPSS apparatus.

The soil samples were sieved into six size-fractions with physical diameters greater than 600 µm, 300 to 600 µm, 150 to 300 µm, 75 to 150 µm, 38 to 75 µm, and less than 38 µm. The 38-µm fraction was further separated by aerodynamic sizing into five size-fractions with aerodynamic diameters less than 1 µm, less than 2.5 µm, less than 5 µm, less than 10 µm, and less than 15 µm. No additional analysis was conducted on the 38-µm to 15-µm particle-size range. Quanterra Environmental Services, Richland Laboratory, analyzed the aerodynamically separated particle samples for plutonium, americium, and uranium using alpha spectroscopy. UNLV's Department of Health Physics analyzed the sieved size-fractions for americium using gamma spectroscopy. Quality assurance/quality control included steps during sieving operations to minimize loss of material during shaking and during transfer from the sieves for weighing and analysis. For filter SSPSS operations and filter analysis quality assurance/quality control steps including simultaneous sampling of two, less than 5-µm size-fraction samples; random weighing of a control set of filters and with other calibration weights; leak tests and flow rate checks of the SSPSS equipment during operation, and a 5 to 10 minute “settling” period before filters were removed to help ensure that no particles remained in suspension.

Aerodynamically separated particles from 15 soil samples (five soil samples at each location) were analyzed by alpha spectroscopy for $^{239+240}$Pu and ^{241}Am. The analyzed activities for $^{239+240}$Pu ranged from 54 to 1,390 pCi/g for location one, from 450 to 9,000 pCi/g for location two, and from 83 to 1,600 pCi/g for location three. The analyzed activities for ^{241}Am ranged from 10 to 254 pCi/g for location one, from 14 to 1,490 pCi/g for location two, and from 14 to 220 pCi/g for location three. Several soil samples had more activity in the smaller size-fractions than in the larger ones, which indicates that radioactive particles were uniformly distributed in the SSPSS suspension chamber during particle separation. The filters were also analyzed for uranium by alpha

spectroscopy. Uranium was found on four samples at low levels: 1.12 to 3.72 pCi/g (0.06 to 0.11 pCi total activity).

For the aerodynamically separated samples, the average ratio of $^{239+240}$Pu to ^{241}Am for all size-fractions was 7.1 with a standard deviation of 10.8, a range of 0.8 to 78.5. The averages for the different size-ranges had slight but not significant differences. At the 95% confidence limit, the range was 4.6 to 9.6. The ratio of 7.1 is lower than the ratio of $^{239+240}$Pu to ^{241}Am found for soil samples at safety test sites on the Tonopah Test Range, consistent with previous work at Project 57.

The activities in the size-intervals for aerodynamic diameters <2.5 μm, from 2.5 to 5 μm, from 5 to 10 μm, and from 10 to 15 μm were computed from the mass and activities on filter samples. The mean activity of $^{239+240}$Pu on the aerodynamically separated samples increased as the particle size increased: 377 pCi/g for the <2.5 μm interval, 592 pCi/g for the 2.5 to 5 μm interval, 1,211 pCi/g for 5 to 10 μm interval, and 1,922 pCi/g for the 10 to 15 μm interval. Paired t-tests show statistically significant increases in $^{239+240}$Pu activity between the <2.5 μm and 2.5-5 μm intervals (at the 3.4% level) and the 2.5-5 μm and 5-10 μm intervals (at the 1.3% level). The increase between the 5-10 μm and 10-15 μm interval had a significance level of 12.5% and would not be considered statistically significant.

The means of the ^{241}Am activities for the aerodynamically separated samples in the <2.5 μm, from 2.5 to 5 μm, from 5 to 10 μm, and from 10 to 15 μm size-intervals were also determined. Although the ^{241}Am activities increased as the particle size increased, no increase was statistically significant.

The relation among ^{241}Am activities of the sieved samples at the three sampling locations and the individual samples was similar to that of the aerodynamically separated samples. The highest activities were found at location two (9 to 2,632 pCi/g); the activity at location three (4 to 1,280 pCi/g) slightly exceeded that at location one (1 to 597 pCi/g). The individual soil samples with the highest alpha activities also had the highest gamma activities; the individual soil samples with the lowest alpha activities also had the lowest gamma activities. A few soil samples had one or more size-intervals with much higher activity than is found in other size-intervals for that sample, which is indicative of the inhomogeneity of the mixture of radionuclides in the sample. However, in general, the results suggest that the plutonium and americium were similarly distributed among different soil particle sizes at the sampling sites.

The mean activity of the sieved samples decreased as the particle size increased for all but the 150 to 300 μm size-interval. Except for one soil sample with a high ^{241}Am activity in the 150 to 300 μm interval, mean activities decreased with increasing particle size for all sizes. Paired t-Tests confirm that the decreases in the means with increasing particle size were highly statistically significant for the <38 μm to 38-75 μm interval (at the 0.0% level), for the 75-15 μm to 150-300 μm interval (at the 0.8% level) and for the 150-300 μm to 300-600 μm interval (at the 0.6% level). The decrease for the 300-600 μm to >600 μm interval was slightly less significant but still at the 3.5% level. The decrease in the mean activity between the 38-75 μm to 75-150 μm interval had a significance level of 7.2%, only slightly statistically significant.

The activities of ^{241}Am in the sieved samples, particularly in the small size-intervals, are higher (range of 8.4 to 2,630 pCi/g) than might be expected given the activities of ^{241}Am and

$^{239+240}$Pu in the aerodynamically separated samples. If the same ratio of about 5 for plutonium to americium as for the aerodynamically separated size-fractions holds for the sieved samples, the plutonium activity in the <38 μm size-range would be from 800 to 13,000 pCi/g. The results of the gamma counting of ^{241}Am and subsequent calculations have been checked and determined to be correct to within the uncertainties of the measurement. A large amount of plutonium associated with particles having aerodynamic diameters greater than 15 μm (a diameter of about 3 μm for a plutonium particle or 5 μm for a plutonium oxide particle) and physical sizes less than 38 μm could result in the high activities in this size range.

The total amount of ^{241}Am in each sieved sample cannot be determined, because the sieved fractions were not weighed when the separations were done. The sieved fractions were not originally intended to be analyzed. The sieved fractions were analyzed only after a gross gamma survey meter indicated relatively high activities in some of the sieved fractions. Previous work at the site by Papelis et al. (1996) indicated slightly more than 50% of the Pu activity is associated with particles less than 38 μm. However, variability between samples was high. In the future, for any work with the SSPSS, the total weight of the soil sample, the weight of the part that is sieved, and the weights of each sieved size-fraction should be determined. The principal value of this would be for a better understanding of activity as a function of soil mass distribution, both in the inhalable and larger soil particle-size ranges.

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ACRONYMS

AD	aerodynamic diameters
Am	americium
DAS	Division of Atmospheric Science
DRI	Desert Research Institute
HEPA	high-efficiency particle arresting
HRC	Harry Reid Center
PD	physical diameters
Pu	plutonium
QES	Quanterra Environ-Services
SOP	standard operating procedure
SSPSS	suspended soil particle sizing system
TTR	Tonopah Test Range
UNLV	University of Nevada, Las Vegas

1.0 INTRODUCTION

As part of the preliminary site characterization conducted for Project 57, soil samples were collected for separation into several size-fractions using the Suspended Soil Particle Sizing System (SSPSS), designed and fabricated by the Division of Atmospheric Sciences (DAS) of the Desert Research Institute (DRI) (Bowen et al., 1998; Chow et al., 1994). Soil samples were collected specifically for separation by the SSPSS at three general locations in the deposited Project 57 plume, the projected radioactivity of which ranged from 100 to 600 pCi/g. The primary purpose in focusing on samples with this level of activity is that it would represent anticipated residual soil contamination levels at the site after corrective actions are completed. Consequently, the results of the SSPSS analysis can contribute to dose calculation and corrective action-level determinations for future land-use scenarios at the site.

Project 57 was a safety test conducted on the Nellis Air Force Range. The purpose of the test was to evaluate the dispersal of plutonium resulting from a chemical explosion of a simulated nuclear device. IT Corporation collected soil samples for the SSPSS analysis as part of preliminary characterization work at the site. The SSPSS operations, consisting of particle separation by sieving and aerodynamic sizing, were performed by personnel of the Harry Reid Center for Environmental Studies (HRC) at the University of Nevada, Las Vegas (UNLV). Sieving was done in the Radiation Protection Laboratory at UNLV. The aerodynamic particle separation was done at the HRC. Quanterra Environmental Services (QES), Richland Laboratory, analyzed the aerodynamically separated particle samples for isotopes of plutonium, americium, and uranium. UNLV's Department of Health Physics analyzed the sieved size-fractions for americium using gamma spectroscopy.

2.0 SOIL SAMPLE COLLECTION

During collection of soil samples for other preliminary site characterization, IT Corporation collected soil samples specifically for the separation of soil particles by the SSPSS. At each sample site, surface vegetation and debris were removed and soil was collected to a depth of approximately 8 cm, placed in a 2-liter container, and marked with a sample identification number. A total of 17 samples were collected at three general locations relative to the estimated plume area shown in Figure 1. The projected activity of the areas of collection was in the range of 100 to 600 pCi/g, the anticipated residual contaminant level range that will remain at the site after corrective action. Higher activity soils were also not considered because of restrictions on the amount of radioactive material that UNLV was licensed to have in its possession. Five soil samples with these activities could be transferred to UNLV at one time for separation. The sample locations and sample identification numbers are listed in Table 1. Two of the samples, PR57-058 and PR57-057, were collected at the same location as samples PR57-035 and PR57-040, respectively, to serve as duplicate samples.

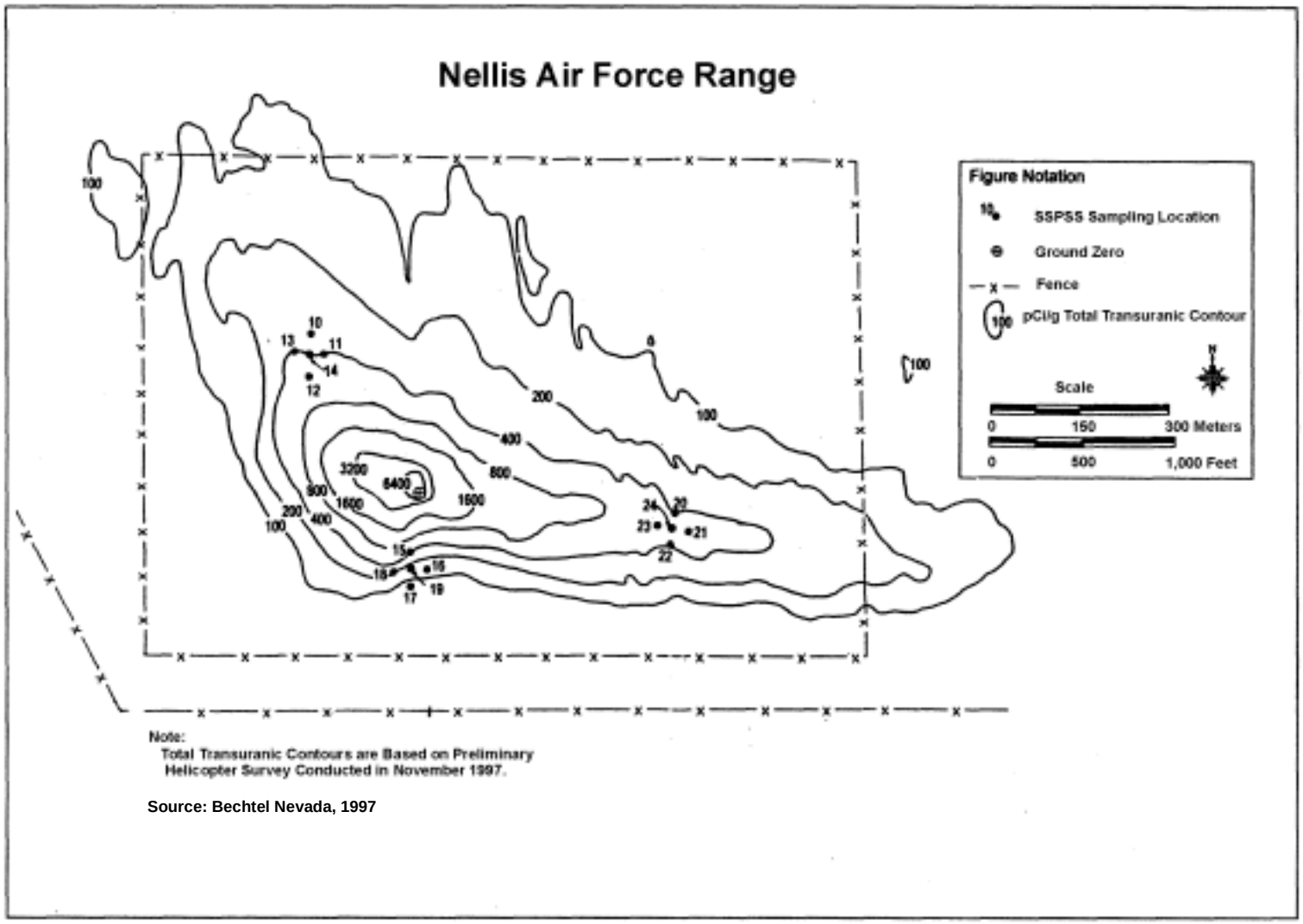

Figure 1. Location of soil samples at Project 57.

Table 1. Project 57 soil samples.

Sample Location	Sample ID	From Ground Zero		Comment
		Bearing Degrees Relative to North	Distance (m)	
Location One				
10	PR57-026	327	324	
11	PR57-027	327	285	
12	PR57-028	318	270	
13	PR57-029	319	318	
14	PR57-030	320	297	
Location Two				
15	PR57-031	187	99	
16	PR57-032	174	129	Not Separated
17	PR57-033	185	159	
18	PR57-034	198	141	
19	PR57-035	186	129	
19	PR57-058	186	129	Duplicate
Location Three				
20	PR57-036	94	438	
21	PR57-037	98	465	
22	PR57-038	102	438	Not Separated
23	PR57-039	98	411	
24	PR57-040	98	438	
24	PR57-057	98	438	Duplicate

3.0 SSPSS OPERATIONS AND ANALYTICAL RESULTS

UNLV received soil samples from IT Corporation in three separate shipments, one for each sampling location. Each set consisted of the five or six samples indicated in Table 1. The soil samples were dried and then sieved into six size-fractions, the smallest of which had particles with physical diameters less than 38 μm. The <38 μm fraction was further separated into five size-fractions with aerodynamic diameters <1, <2.5, <5, <10, and <15 μm, respectively. The samples were received, transported, handled, and stored in accordance with the Radiation Safety Standard Operating Procedure (SOP) for the Soil Analyses Laboratory and SSPSS. Training of personnel was performed in accordance with this SOP. Figure 2 contains a flow diagram of the operations associated with the SSPSS and analytical operations that are described in this section. In the following discussion, particle size-fractions are determined by the method of separation: physical diameters (PD) for the sieving process and aerodynamic diameters (AD) referenced to a spherical particle with a density of 1 g/cm^3.

3.1 Sieving Operations

Sieving operations were performed in a large laboratory hood in UNLV's Radiation Protection Laboratory. The hood was equipped with a high-efficiency particle arresting (HEPA) filter on the exhaust to prevent possible release of contaminated dust to the atmosphere. Prior to sieving, the bulk soil samples were dried at 110°C for several hours. For each soil sample, approximately half a kilogram of soil was placed in the top sieve of the stack. The sieve stack was shaken for 20 minutes to separate the soil into six size-fractions with physical diameters greater than 600 μm, 300 to 600 μm, 150 to 300 μm, 75 to 150 μm, 38 to 75 μm, and less than 38 μm. The five larger size-fractions were placed in labeled bags for possible further analysis. The <38 μm size-fraction was placed in a 50-ml container and taken to the suspension area for aerodynamic separation to smaller size-fractions. The shaker stakes were locked together during the sieving operation to minimize the potential for fine particles to escape. In addition, the stake was observed during the entire 6 to 15 minute shaking operation. No escape of fines was observed.

3.2 Suspension Operations

Suspension operations were performed in a small laboratory room in the HRC. Inside the laboratory, a secondary containment structure was constructed of PVC tubing and polyethylene sheeting to contain soil particles that might be released during operations of the apparatus. The most likely time for a release of particles to the air was during the cleanup after the separation process. A vacuum cleaner with a HEPA filter was used to remove soil that fell out in the suspension chamber. An air-cleaning device with HEPA filter was installed in the secondary containment area to collect dust released during cleanup operations. A HEPA filter was also installed on the exhaust fan outlet from the room to prevent release of contaminated particles to the atmosphere.

For each soil sample, a portion of the final sieved product (<38 μm fraction) was separated aerodynamically into five size-fractions with aerodynamic diameters <1 μm, <2.5 μm, <5 μm, <10 μm, and <15 μm, respectively. The SSPSS separation apparatus is shown schematically in Figure 3. The <38 m sample was introduced to the suspension chamber through the suspension flask. The suspended particles separated into different size-fractions by the impactors specific to

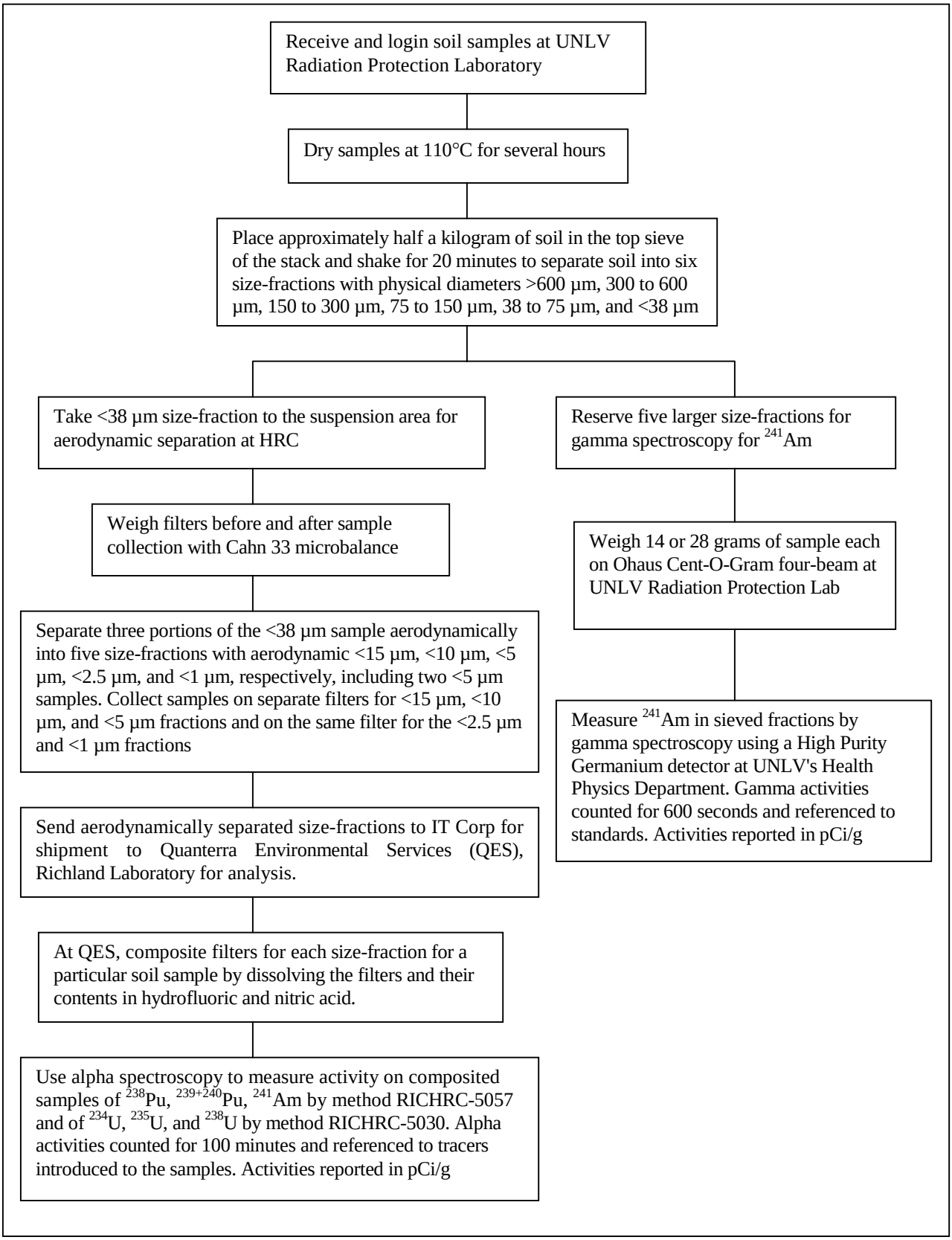

Figure 2. Operations associated with the SSPSS and analytical operations.

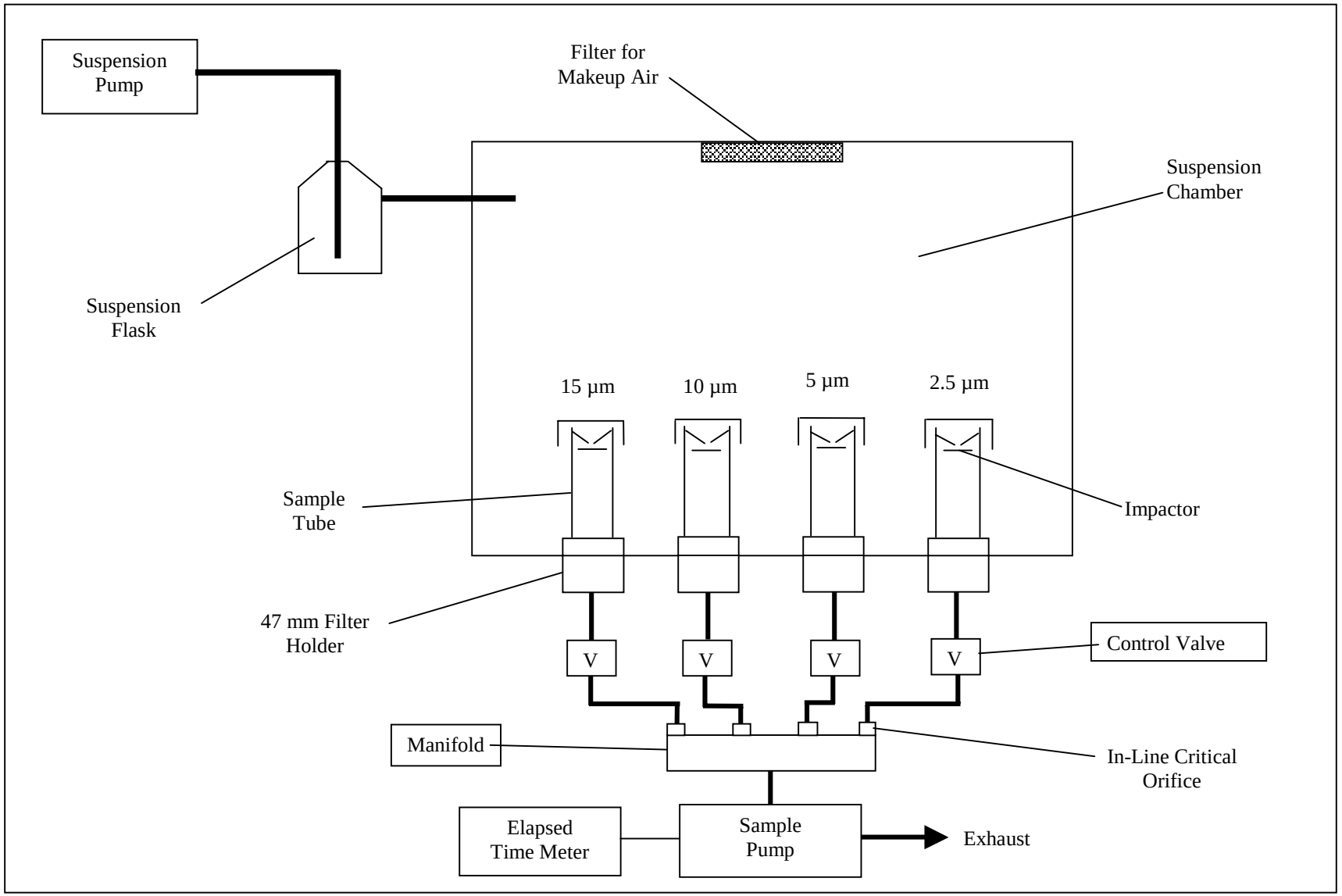

Figure 3. Schematic sketch of SSPSS suspension equipment.

each size and were collected on filters for subsequent analysis. A second sample in the <5 μm size-fraction was also collected as a check of the homogeneity of the suspended particles. To collect the amount of mass required for the analysis of plutonium, three suspensions were performed for each soil sample. The <5 μm, <10 μm and <15 μm size-fractions were collected on separate filters for each suspension, so as not to overload the filters and impaction plates. For all but two soil samples, the <1 μm and <2.5 μm size-fractions were collected on single filters, because of the small amount of mass collected for those sizes. The filters were weighed before and after the soil collection with a Cahn Model 33 microbalance to determine the amount of suspended soil collected. A control set of filters and calibration weights were randomly re-weighed as a quality control procedure. This was done in place of making multiple measurements of the filters to minimize handling of the filters during which material could be lost. In addition, particularly for the benefit of the finest filter fractions, a 5 to 10 minute "settling" period was used after operation of the SSPSS apparatus before the filters were removed to help ensure that no fine particles remained in suspension. In addition, leak tests and flow rate checks, in accordance with the DRI SOP, were performed during the SSPSS operation.

The <1 μm samples for the Project 57 samples contained higher than expected masses. The average ratio of mass on the <1 μm samples to mass on the <2.5 μm samples was 0.66 ± 0.08; i.e., two thirds of the <2.5 μm size-fraction would have consisted of particles smaller than <1 μm. Other measurements have found that airborne particles originating from soil have less than 10% of the total mass in the <1 μm and smaller size-fraction (Friedlander, 1977; Whitby, 1978). Airborne particles that originate as suspended soil particles typically have a lognormal mass distribution with a maximum near 9 μm and geometric standard deviation of about 5 μm. The mass in the <1 μm size-fraction for such a distribution would be about 7% of the mass in the <2.5 μm size-fraction, or much less than the 66% found for the current measurements. The processes used in the SSPSS to separate particles, while not exactly the same as those that would generate airborne particles by natural processes, would not have modified the soil sample so much as to result in the measured abundance of mass in the <1 μm size-range. As a consequence, the results of <1 μm samples were combined with the <2.5 μm fraction for analysis. The <1 μm samples will not be considered further during this report.

The aerodynamically separated samples are listed in Tables 2 through 4. Also included are filter numbers and individual masses and the composite sets and total mass of all the filters for a particular size-fraction for each sample. The samples labeled 5A are those for the second <5 μm size-fraction.

3.3 Radiological Results

Both the sieved fractions and the aerodynamically separated fractions were analyzed for radionuclides. The following section describes the two types of analyses performed.

Aerodynamically Separated Fractions – Alpha Spectroscopy

The aerodynamically separated samples were analyzed for plutonium ($^{239+240}$Pu and ^{238}Pu), americium (^{241}Am), and uranium (^{234}U, ^{235}U, and ^{238}U) by QES, Richland Laboratory. After aerodynamic separation, and sample collection and weighing at UNLV, the sample filters were sent to IT Corporation for shipment to QES. All soil samples had three filters for each size-fraction at 5 μm and larger. A few of the smaller size-fractions also had multiple filters but most samples consisted of a single filter. At QES, the filters for each size-fraction for a particular soil sample were

Table 2. Mass on aerodynamically separated size-fractions – location one.

Soil Sample ID	Size-Fraction, Aerodynamic Diameter, µm	Filter 1		Filter 2		Filter 3		Sum Mass Mg
		ID	Mass, mg	ID	Mass, mg	ID	Mass, mg	
PR57-026	<1	PR57001	3.686	-	-	-	-	3.686
	<2.5	PR57002	2.060	PR57007	1.461	PR57012	3.120	6.641
	<5	PR57003	11.392	PR57008	8.364	PR57013	2.803	22.559
	<5A	PR57004	12.645	PR57009	8.694	PR57014	15.039	36.378
	<10	PR57005	21.204	PR57010	14.938	PR57015	33.136	69.278
	<15	PR57006	25.991	PR57011	17.497	PR57016	26.386	69.874
PR57-027	<1	PR57034	2.160	-	-	-	-	2.160
	<2.5	PR57035	3.827	-	-	-	-	3.827
	<5	PR57036	6.812	PR57040	6.435	PR57044	5.507	18.754
	<5A	PR57037	7.680	PR57041	7.773	PR57045	7.048	22.501
	<10	PR57038	15.941	PR57042	14.956	PR57046	8.974	39.871
	<15	PR57039	22.577	PR57043	19.607	PR57047	15.601	57.785
PR57-028	<1	PR57048	5.436	-	-	-	-	5.436
	<2.5	PR57049	9.881	-	-	-	-	9.881
	<5	PR57050	17.617	PR57054	12.938	PR57058	10.476	41.031
	<5A	PR57051	18.584	PR57055	12.296	PR57059	10.880	41.760
	<10	PR57052	33.033	PR57056	26.983	PR57060	23.669	83.685
	<15	PR57053	43.453	PR57057	31.364	PR57061	19.889	94.706
PR57-029	<1	PR57018	3.209	PR57024	2.115	-	-	5.324
	<2.5	PR57019	1.552	PR57025	8.092	-	-	9.644
	<5	PR57020	8.370	PR57026	9.329	PR57030	11.055	28.754
	<5A	PR57021	9.786	PR57027	16.726	PR57031	12.515	39.027
	<10	PR57022	19.410	PR57028	23.914	PR57032	21.310	64.634
	<15	PR57023	14.486	PR57029	2.795	PR57033	30.244	47.525
PR57-030	<1	PR57062	4.153	-	-	-	-	4.153
	<2.5	PR57063	5.188	-	-	-	-	5.188
	<5	PR57064	8.183	PR57068	9.641	PR57072	6.995	24.819
	<5A	PR57065	9.769	PR57069	11.370	PR57073	10.732	31.871
	<10	PR57066	15.375	PR57070	21.440	PR57074	16.830	53.645
	<15	PR57067	19.128	PR57071	24.752	PR57075	24.440	68.320

Table 3. Mass on aerodynamically separated size-fractions – location two.

Soil Sample ID	Size-Fraction, Aerodynamic Diameter, μm	Filter 1		Filter 2		Filter 3		Sum Mass Mg
		ID	Mass, mg	ID	Mass, mg	ID	Mass, mg	
PR57-031	<1	PR57076	4.567	-	-	-	-	4.567
	<2.5	PR57077	7.205	-	-	-	-	7.205
	<5	PR57078	9.782	PR57082	6.746	PR57086	17.937	34.465
	<5A	PR57079	9.683	PR57083	7.957	PR57087	18.167	35.807
	<10	PR57080	22.430	PR57084	12.280	PR57088	36.114	70.824
	<15	PR57081	27.215	PR57085	19.002	PR57089	43.109	89.326
PR57-033	<1	PR57090	7.508	-	-	-	-	7.508
	<2.5	PR57091	9.886	-	-	-	-	9.886
	<5	PR57092	13.538	PR57096	10.908	PR57100	12.597	37.043
	<5A	PR57093	13.914	PR57097	12.950	PR57101	13.827	40.691
	<10	PR57094	29.579	PR57098	23.644	PR57102	24.870	78.093
	<15	PR57095	40.264	PR57099	33.422	PR57103	33.205	106.891
PR57-034	<1	PR57104	5.432	-	-	-	-	5.432
	<2.5	PR57105	8.262	-	-	-	-	8.262
	<5	PR57106	9.053	PR57110	13.795	PR57114	13.193	36.041
	<5A	PR57107	9.227	PR57111	15.509	PR57115	15.427	40.163
	<10	PR57108	20.031	PR57112	31.901	PR57116	41.339	93.271
	<15	PR57109	27.350	PR57113	43.719	PR57117	28.551	99.620
PR57-035	<1	PR57118	7.966	-	-	-	-	7.966
	<2.5	PR57119	11.256	-	-	-	-	11.256
	<5	PR57120	20.330	PR57124	12.798	PR57128	15.874	49.002
	<5A	PR57121	21.784	PR57125	13.449	PR57129	16.977	52.210
	<10	PR57122	38.859	PR57126	25.984	PR57130	33.404	98.247
	<15	PR57123	53.334	PR57127	37.796	PR57131	42.263	133.393
PR57-058	<1	PR57132	5.780	-	-	-	-	5.780
	<2.5	PR57133	8.734	-	-	-	-	8.734
	<5	PR57134	14.290	PR57138	10.658	PR57142	9.014	33.962
	<5A	PR57135	15.487	PR57139	13.672	PR57143	11.154	40.313
	<10	PR57136	31.640	PR57140	23.237	PR57144	24.097	78.974
	<15	PR57137	38.912	PR57141	36.774	PR57145	36.941	112.627

Table 4. Mass on aerodynamically separated size-fractions – location three.

Soil Sample ID	Size-Fraction, Aerodynamic Diameter, μm	Filter 1		Filter 2		Filter 3		Sum Mass Mg
		ID	Mass, mg	ID	Mass, mg	ID	Mass, mg	
PR57-036	<1	PR57146	6.804	-	-	-	-	6.804
	<2.5	PR57147	8.838	-	-	-	-	8.838
	<5	PR57148	13.314	PR57152	11.380	PR57156	12.936	37.630
	<5A	PR57149	13.813	PR57153	13.061	PR57157	14.743	41.617
	<10	PR57150	30.777	PR57154	26.636	PR57158	27.280	84.693
	<15	PR57151	36.244	PR57155	35.841	PR57159	33.042	105.127
PR57-037	<1	PR57160	5.593	-	-	-		5.593
	<2.5	PR57161	8.824	-	-	-	-	8.824
	<5	PR57162	14.177	PR57166	14.976	PR57170	12.546	41.699
	<5A	PR57163	16.816	PR57167	16.723	PR57171	13.497	47.036
	<10	PR57164	30.761	PR57168	31.784	PR57172	25.185	87.730
	<15	PR57165	39.785	PR57169	38.551	PR57173	30.441	108.777
PR57-039	<1	PR57174	8.689	-	-	-	-	8.689
	<2.5	PR57175	12.892	-	-	-	-	12.892
	<5	PR57176	15.333	PR57180	20.728	PR57184	19.940	56.001
	<5A	PR57177	18.635	PR57181	22.787	PR57185	21.288	62.710
	<10	PR57178	34.762	PR57182	43.598	PR57186	39.807	118.167
	<15	PR57179	50.015	PR57183	51.730	PR57187	49.705	151.45
PR57-040	<1	PR57188	4.652	-	-	-	-	4.652
	<2.5	PR57189	7.508	-	-	-	-	7.508
	<5	PR57190	9.615	PR57194	12.258	PR57198	10.265	32.138
	<5A	PR57191	11.140	PR57195	13.540	PR57199	11.948	36.628
	<10	PR57192	21.428	PR57196	25.174	PR57200	26.261	72.863
	<15	PR57193	31.132	PR57197	35.611	PR57201	34.672	101.415
PR57-057	<1	PR57202	4.957	-	-	-	-	4.957
	<2.5	PR57203	7.152	-	-	-	-	7.152
	<5	PR57204	10.527	PR57208	14.076	PR57212	8.780	33.383
	<5A	PR57205	11.428	PR57209	14.236	PR57213	9.651	35.315
	<10	PR57206	22.959	PR57210	35.308	PR57214	18.335	76.602
	<15	PR57207	31.530	PR57211	40.791	PR57215	27.279	99.600

composited by dissolving the filters and their contents in hydrofluoric and nitric acid. Alpha spectroscopy was used to quantify the americium, plutonium isotopes, and uranium isotopes on the composited filters. Method RICHRC-5057 was used for ^{241}Am, $^{239+240}$Pu, and ^{238}Pu; method RICHRC-5030 was used for ^{234}U, ^{235}U, and ^{238}U. In all cases, the analyses were done at reduced volume because of the relatively small amount of mass on the filters. The alpha activities were counted for 100 minutes and referenced to tracers introduced to the samples. Activities were reported in pCi/g.

A total of 15 sets of filters were analyzed by alpha spectroscopy. The resulting activities in pCi/g and pCi/filter are given in Tables 5 through 7 for $^{239+240}$Pu and ^{241}Am for the 2.5 μm and larger aerodynamically separated size-fractions. Those samples with activity below detection limits (one $^{239+240}$Pu and two ^{241}Am) are enclosed in parentheses on the Tables. Uranium was measured above detection limits in four samples as noted in footnotes in the Tables. The uranium activities will not be discussed further because of their low values and few samples on which they were found above detection limits. The activities are listed for particles in the composited filter sample with sizes less than the aerodynamic diameter listed at the top of each column. The mass presented on Tables 2, 3, and 4 for each particle size-range represents the mass collected on that filter as well as the cumulative mass of the finer soil particle fractions. In addition, the mass percentages of each particle size-fraction as a function of the total mass of each sample are provided as an additional means of evaluation of distribution of total inhalable particles and contaminants.

In a well-mixed suspension of particles in which the radioactive particles are uniformly distributed throughout the suspension chamber, the total activity on each filter should decrease as the particle size-fraction decreases, because each smaller fraction would contribute the same activity to larger fractions. Several samples, however, had more activity in the smaller size-fraction than in the larger ones. This could be the result of an inhomogeneous mixture of radioactive particles in the suspension, or for size-fractions with low mass, a single “hot particle” with attached plutonium or americium isotopes attached could similarly affect the results. The activities that were less than those for a smaller size range are underlined in the Tables. The inhomogeneity is particularly clear in several of the duplicate 5 μm samples. The subsequent analysis has used the average of the 5 μm samples.

The overall activity of soil particles in the inhalable size range of the three areas where samples were collected can be determined from the activities on the 15 μm samples. These activities are also a measure of the total activity of particles in the inhalable size-ranges. The activities of $^{239+240}$Pu in the 15-μm samples range from 82 to 692 pCi/g for location one, from 818 to 2,940 pCi/g for location two, and from 189 to 1,600 pCi/g for location three.

The ratios of $^{239+240}$Pu to ^{241}Am have been computed for each size-fraction for each soil sample and are given in Table 8. For all samples except those with activities less than the detection limit, the average ratio for all the size-fractions was 7.1, with a standard deviation of 10.8 and a range of 0.8 to 78.5. The 95% confidence interval was 4.6 to 9.6. The averages for the different size-ranges had slight but not significant differences. The average ratio is reduced to 5.3 if the two large ratios, which appear to have much lower americium values than do other samples, are

Table 5. Radioactivity on aerodynamically separated size-fractions – location one.

Soil Sample ID	Size-Fraction, Aerodynamic Diameter, µm	Activity for $^{239+240}$Pu		Activity for ^{241}Am	
		PCi/g	PCi/Sample	PCi/g	PCi/Sample
PR57-026[a]	<2.5	65.4	0.4	*(6.26)*[d]	*(0.04)*
	<5	94.7	2.1	16.2	0.4
	<5A	98.1	3.6	15.9	0.6
	<10	261	18.1	43.9	3.0
	<15[a]	109	7.6[e]	9.5	0.7
PR57-027[b]	<2.5	162	0.6	50	0.2
	<5[b]	193	3.6	38.7	0.7
	<5A	1390	31.3	254	5.7
	<10	281	11.2	71.4	2.9
	<15	330	19.1	52.9	3.1
PR57-028[c]	<2.5	380	3.6	57.8	0.6
	<5[c]	314	12.9	53.6	2.2
	<5A	321	13.4	61.9	2.6
	<10	838	70.2	171	14.3
	<15	522	49.4	87.6	8.3
PR57-029	<2.5	*(6.54)*	0.1	88.1	0.9
	<5	54.2	1.6	*(2.44)*	*(0.07)*
	<5A	70.1	2.7	11.5	0.5
	<10	139	9.0	45.4	2.9
	<15	81.6	3.9	16.1	0.8
PR57-030	<2.5	158	0.8	47.1	0.2
	<5	259	6.4	60.5	1.5
	<5A	379	12.1	67.7	2.2
	<10	439	23.6	92.3	5.0
	<15	692	47.3	120	8.2

[a] ^{234}U found above detection limit in <15 µm fraction: 1.52 pCi/g, 0.11 pCi/sample.
[b] ^{238}U found above detection limit in <5 µm fraction: 3.66 pCi/g, 0.070 pCi/sample.
[c] ^{238}U found above detection limit in <5 µm fraction: 1.60 pCi/g, 0.066 pCi/sample.
[d] Activities in *( )* are below detection limits for ^{241}Am or $^{239+240}$Pu.
[e] Underlined activity indicates value less than in next smaller size-fraction.

Table 6. Radioactivity on aerodynamically separated size-fractions – location two.

Soil Sample ID	Size-Fraction, Aerodynamic Diameter, µm	Activity for $^{239+240}$Pu		Activity for ^{241}Am	
		PCi/g	pCi/Sample	pCi/g	pCi/Sample
PR57-031	<2.5	1300	9.4	222	1.6
	<5	3140	108.2	316	10.9
	<5A	1500	53.7	314	11.4
	<10	3870	274.1	738	52.3
	<15	2940	262.6[b]	501	44.8
PR57-033	<2.5	630	6.2	86.6	0.9
	<5	562	20.8	101	3.7
	<5A	757	30.8	155	6.3
	<10	1170	91.4	203	15.9
	<15	818	87.4	125	13.4
PR57-034	<2.5	452	3.7	55.4	0.5
	<5	517	18.6	90.3	3.3
	<5A	389	15.6	218	8.8
	<10	522	48.7	175	16.3
	<15	834	83.1	13.8	1.4
PR57-035[a]	<2.5	512	5.8	660	7.4
	<5[a]	9030	442.5	1490	73.0
	<5A	706	36.9	519	27.1
	<10	1480	145.4	447	43.9
	<15	1860	248.1	23.7	3.5
PR57-058	<2.5	1070	9.3	267	2.3
	<5	1610	54.7	316	10.7
	<5A	3930	158.4	790	31.9
	<10	1260	99.5	195	15.4
	<15	2860	322.1	593	66.8

[a] ^{238}U found above detection limit in <5 µm fraction: 1.12 pCi/g, 0.055 pCi/sample.
[b] Underlined activity indicates value less than in next smaller size-fraction.

Table 7. Radioactivity on aerodynamically separated size-fractions – location three.

Soil Sample ID	Size-Fraction, Aerodynamic Diameter, µm	Activity for $^{239+240}$Pu		Activity for ^{241}Am	
		PCi/g	pCi/Sample	pCi/g	pCi/Sample
PR57-036	<2.5	315	2.8	54.1	0.5
	<5	731	27.5	156	5.6
	<5A	621	25.8	119	5.0
	<10	1340	113.5	217	18.4
	<15	1600	168.2	220	23.1
PR57-037	<2.5	166	1.5	38.1	0.3
	<5	233	9.7	43.8	1.8
	<5A	119	5.6	34.4	1.6
	<10	434	38.1	69.4	6.1
	<15	436	47.4	89.9	9.8
PR57-039	<2.5	82.7	1.1	14	0.2
	<5	88.6	5.0	17.6	1.0
	<5A	105	6.6	21.1	1.3
	<10	183	21.6	35.9	4.2
	<15	189	28.6	34.3	5.2
PR57-040	<2.5	190	1.4	23.3	0.2
	<5	270	8.7	51	1.6
	<5A	260	9.5	44.5	1.6
	<10	623	45.4	112	8.2
	<15	487	49.4	90.3	9.2
PR57-057	<2.5	161	1.2	32.3	0.2
	<5	250	8.3	54.4	1.8
	<5A	211	7.5	46.1	1.6
	<10	347	26.6	77.3	5.9
	<15	350	34.9	75.2	7.5

[a] No samples with Uranium above detection limits.

Table 8. Ratio of $^{239+240}$Pu to ^{241}Am activity for aerodynamic size-fractions.

Soil Sample ID	Activity of ^{241}Am in pCi/g				
	Aerodynamic Diameter in µm				
	d < 2.5	d < 5		d < 10	d < 15
		First	Second		
PR57-026	*(10.4)*[a]	5.8	6.2	5.9	11.5
PR57-027	3.2	5.0	5.5	3.9	6.2
PR57-028	6.6	5.9	5.2	4.9	6.0
PR57-029	*(0.1)*[a]	*(22.2)*[a]	6.1	3.1	5.1
PR57-030	3.4	4.3	5.6	4.8	5.8
PR57-031	5.9	9.9	4.8	5.2	5.9
PR57-033	7.3	5.6	4.9	5.8	6.5
PR57-034	8.2	5.7	1.8	3.0	60.4
PR57-035	0.8	6.1	1.4	3.3	78.5
PR57-058	4.0	5.0	5.1	6.5	4.8
PR57-036	5.8	4.7	5.2	6.2	7.3
PR57-037	4.4	5.3	3.5	6.3	4.8
PR57-039	5.9	5.0	5.0	5.1	5.5
PR57-040	8.2	5.3	5.8	5.6	5.4
PR57-057	5.0	4.6	4.6	4.5	4.7
Average	5.3	5.6	4.7	4.9	14.6
Std Dev	21	1.4	1.4	1.2	22.6
Maximum	8.2	9.9	6.2	6.5	78.5
Minimum	0.8	4.3	1.4	3.0	4.7
Number	13	14	15	15	15
All Size Ranges	Average	7.1	95% Confidence Limits		
	Std Dev	10.8	+95%		9.6
	Number	72	-95%		4.6

[a] Ratios in *( )* have ^{241}Am or $^{239+240}$Pu below detection limits – not used in averages.

not included in the average. Even the higher ratio of 7.1 is approximately three to four times lower than the ratio of $^{239+240}$Pu to ^{241}Am found for soil samples at safety test sites on the Tonopah Test

Range (TTR). Differences in the ratio of these isotopes at the Project 57 site compared to the TTR sites have been noted in previous studies, including Gilbert et al. (1975) and Kroodsma (1994), although methods used and ratios calculated varied from one study to another. Gilbert et al. (1975) calculated ratios based on over 150 samples collected over the entire aerial extent of the Project 57 plume, with samples collected to a depth of approximately 5 cm. Plutonium to americium ratios in Gilbert et al. (1975) for Project 57 were "9 to 10" versus "22 to 26" at the TTR sites.

The activities in the size-intervals for aerodynamic diameters <2.5 μm, from 2.5 to 5 μm, from 5 to 10 μm, and from 10 to 15 μm have been computed from the mass and activities on filter samples. The activity in a particular size-interval was computed from

$$\text{Activity} = \frac{a_j M_j - a_{j-1} M_{j-1}}{M_j - M_{j-1}}$$

where a_j is activity in the jth size-fraction, M_j is mass in the jth size-fraction, and j-1 is the next smaller size-interval. The computation assumes that the activity and mass associated with particles on the filters for the smaller size-fraction are similar to the activity and mass that size-fraction contributes to the filters for the larger size-fraction. The activities in the size-intervals are given in Table 9 for $^{239+240}$Pu and in Table 10 for ^{241}Am. The smallest size-interval considered here has a diameter less than 2.5-μm computations. These filters had masses for the <10 μm and <15 μm size-fractions that were nearly equal, while the measured activity was higher on the <10 μm filters than on the <15 μm filters. Negative values for the 10- to15-μm interval may be attributed to the inhomogeneous mixture of soil and radionuclides that were introduced to the suspension chamber, errors in the measurement of small amounts of radioactivity, and the possible effects of a "hot particle" on activity distribution, particularly for filters where the mass and total activity were low.

The data indicate that there is a correlation between the aerodynamically separated particle size-fractions and the activity on them, although it is statistically more significant for plutonium than americium isotopes. The means of the $^{239+240}$Pu activities have been computed for the samples that did not have negative values and are included in Table 9. The mean activity increased as the particle size increased. The paired t-Tests in Table 11 show statistically significant increases in activity between the <2.5 μm and 2.5-5 μm intervals (at the 3.4% level) and the 2.5-5 μm and 5-10 μm intervals (at the 1.3% level). The increase between the 5-10 μm and 10-15 μm interval had a significance level of 12.5% and would not be considered statistically significant. The means of the ^{241}Am activities have been computed for the seven sets that did not have negative values and are included in Table 10. The ^{241}Am activities also tended to increase as the particle size increased, but, as shown in Table 11, no increase was statistically significant.

The duplicate soil samples resulted in some differences in activity, although the comparison between PR57-040 and its duplicate PR57-057 was better than the comparison between PR57-035 and PR57-058. The average percent difference between the $^{239+240}$Pu activity on the composite filters for PR57-040 and PR57-057 was -7% with a standard deviation of 52%. A paired t-Test for these samples gave a small t-Statistic of 0.761 for a difference that would be statistically significant at the 48% level, which shows no significant difference between the PR57-040 and PR57-057 samples. With the two largest activities on the 5 μm filters not included, the average percent difference between the $^{239+240}$Pu activity on the composite filters for PR57-035 and PR57-058 was

31% with a standard deviation of 47%. A paired t-Test for these samples gave a t-Statistic of 1.514 for a difference that would have been statistically significant at the 21% level, which shows the samples were different but at a low statistical level. The differences in the duplicate samples are one evidence of the extreme variability in the distribution of radionuclides within safety test contaminant plumes.

Table 9. Activity of $^{239+240}$Pu for aerodynamic size-fractions.

Soil Sample ID	Activity of $^{239+240}$Pu in pCi/g			
	Aerodynamic Diameter in μm			
	d < 2.5	2.5 < d < 5	5 < d < 10	10 < d < 15
Location One				
PR57-026	65	106	383	-17559
PR57-027	162	201	359	439
PR57-028	380	298	1348	-1878
PR57-029	7	86	222	298
PR57-030	158	364	565	1617
Location Two				
PR57-031	1300	2503	5411	-620
PR57-033	630	676	1671	-137
PR57-034	452	449	572	5418
PR57-035	512	759	2358	2922
PR57-058	1070	1797	996	6615
Location Three				
PR57-036	315	776	1926	2678
PR57-037	166	174	702	444
PR57-039	83	101	270	210
PR57-040	190	286	943	140
PR57-057	161	248	442	360
Summary for Locations without Negative Activities				
Average	377	592	1211	1922
Std Dev	375	699	1330	2269
Number	15	15	15	11

Table 10. Activity of ^{241}Am for aerodynamic size-fractions.

Soil Sample ID	Activity of ^{241}Am in pCi/g			
	Aerodynamic Diameter in µm			
	d < 2.5	2.5 < d < 5	5 < d < 10	10 < d < 15
Location One				
PR57-026	6	19	65	-3989
PR57-027	50	203	-18	12
PR57-028	58	58	282	-546
PR57-029	88	-14	97	127
PR57-030	47	69	123	221
Location Two				
PR57-031	222	339	1155	-406
PR57-033	87	144	276	-87
PR57-034	55	101	228	-2354
PR57-035	660	480	365	-1160
PR57-058	267	333	104	1527
Location Three				
PR57-036	54	160	286	232
PR57-037	38	39	101	175
PR57-039	14	21	53	29
PR57-040	23	54	170	35
PR57-057	32	55	99	68
Summary for Locations without Negative Activities				
Average	113	148	243	302
Std Dev	168	142	280	501
Number	15	14	14	8

Table 11. t-Test for activity of $^{239+240}$Pu and ^{241}Am for aerodynamic size-fractions. Comparisons of samples without negative activities.

	Comparison Groups Aerodynamic Diameter in μm		
	<2.5 vs. 2.5 to 5	2.5 to 5 vs. 5 to 10	5 to 10 vs. 10 to 15
$^{239+240}$Pu			
Degrees of freedom	14	14	10
t-Statistic for paired samples	2.342	2.846	1.673
Probability of rejecting null hypothesis	0.034	0.013	0.125
^{241}Am			
Degrees of freedom	13	12	7
t-Statistic for paired samples	1.594	1.650	0.957
Probability of rejecting null hypothesis	0.135	0.125	0.370

Sieved Size Fractions - Gamma Spectroscopy

The sieved fractions were analyzed by gamma spectroscopy for ^{241}Am at UNLV's Health Physics Department with a gamma radiation counter using a High Purity Germanium detector. A portion (14 or 28 grams) of each sieved fraction was weighed on an Ohaus Cent-O-Gram four-beam, hanging-pan balance that was located in the hood in the Radiation Protection Laboratory. Each portion was placed in a plastic dish that was covered, sealed, and transported to the Health Physics Department. The quantity of mass and the dish size were chosen to fit into the counter and to give the proper depth of sample for optimal counting. The duration of most counting periods was 600 seconds with a few samples counted for longer periods. The detected radionuclides included ^{241}Am in all samples and ^{224}Ra and ^{40}K in a few samples. Only the values for ^{241}Am are presented and analyzed herein.

Table 12 lists the ^{241}Am activities in the sample intervals for physical diameters <38 μm, 38 to 75 μm, 75 to 150 μm, 150 to 300 μm, 300 to 600 μm, and >600 μm. The relation among the sieved samples from the three locations and the individual samples was similar to that of the aerodynamically separated samples. The highest activities were found for location two; the activity at location three slightly exceeded that at location one. The individual soil samples with the highest alpha activities also had the highest gamma activities; the individual soil samples with the lowest alpha activities also had the lowest gamma activities. A few soil samples had one or more size-intervals with much higher activity than is found in other size-intervals for the sample, which may be indicative of the inhomogeneity of the mixture of radionuclides in some of the samples. However, in general, the results suggest from the sieved fractions that the plutonium and americium show similar distributions as a function of particle size and that there has not been significant relocation of one versus the other over time.

The mean activity for the sieved fractions decreased with increasing particle size for all but the 150 to 300 μm size interval. One soil sample, PR57-058, had a high ^{241}Am activity in the 150 to 300 μm range. With the PR57-058 sample not included in the averages, the mean activity decreased with increasing particle size for all sizes. Paired t-Tests confirm that, without PR57-058, the decreases in the means with increasing particle size were highly statistically significant for the <38 μm to 38-75 μm interval, for the 75-15 μm to 150-300 μm interval, and for the 150-300 μm to 300-600 μm interval. The decrease for the 300-600 μm to >600 μm interval was slightly less significant but still at the 3.5% level. The decrease in the mean activity between the 38 to 75 μm to 75 to 150 μm interval had a significance level of 7.2%, only slightly statistically significant.

The total amount of ^{241}Am in each sieved sample cannot be determined, because the sieved fractions were not weighed when the separations were done. Similarly, the total activity of the soil samples cannot be determined, particularly since plutonium analysis was not done on them. The sieved fractions were not originally intended to be analyzed. The sieved fractions were analyzed only after a gross gamma survey meter indicated relatively high activities in some of the sieved fractions. Work by Papelis et al. (1996) on six Project 57 samples used to evaluate soil volume reduction technologies showed an average of 54.9% of the Pu activity is associated with particles less than 38 μm. However, the standard deviation of 44.1 indicates the variability between samples.

Table 12. Activity of ^{241}Am for sieved size-fractions.

Soil Sample ID	Activity of ^{241}Am in pCi/g					
	Physical Diameter in μm					
	< 38	38<d<75	75<d<150	150<d<300	300<d<600	d>600
Location One						
PR57-026	423	189	44	15	9	3
PR57-027	323	492	107	34	12	6
PR57-028	551	570	71	31	29	3
PR57-029	279	223	15	7	7	1
PR57-030	390	597	42	14	22	4
Location Two						
PR57-031	1491	1056	421	398	98	120
PR57-033	860	147	151	76	39	9
PR57-034	701	272	126	128	23	18
PR57-035	1328	204	163	192	50	17
PR57-058	2632	508	183	1735	81	17
Location Three						
PR57-036	1280	502	298	39	42	8
PR57-037	565	374	86	23	18	6
PR57-039	177	339	124	18	12	13
PR57-040	789	445	132	27	23	4
PR57-057	325	474	60	23	15	4
Summary for All Locations						
Average	808	426	135	184	32	15
Standard Deviation	649	228	106	441	27	29
Number	15	15	15	15	15	15

Table 13. t-Test for activity of ^{241}Am for sieved size-fractions.

	Comparison Groups Physical Diameter in μm				
	38 to 75 vs. <38	75 to 150 vs. 38 to 150	150 to 300 vs. 75 to 150	300 to 600 vs. 150 to 300	>600 vs. 300 to 600
All Samples					
Degrees of freedom	14	14	14	14	14
t-Statistic for paired samples	2.376	6.165	0.450	1.390	3.256
Probability of rejecting null hypothesis	0.032	0.000	0.659	0.186	0.006
Without Sample Number 58					
Degrees of freedom	13	13	13	13	13
t-Statistic for paired samples	2.360	5.695	3.145	1.955	3.248
Probability of rejecting null hypothesis	0.035	0.000	0.008	0.072	0.006

4.0 CONCLUSIONS

Soil samples were collected specifically for separation by the SSPSS at three general locations in the deposited Project 57 plume, the projected radioactivity of which ranged from 100 to 600 pCi/g. Samples were collected from this range of the plume because it represents what are anticipated to be residual contaminant levels after corrective action at the site. As a consequence, results of the SSPSS analysis can be used for dose-level and corrective action-level determinations for future land-use scenarios. Higher activity soils were also not considered because of restrictions on the amount of radioactive material that UNLV was licensed to have in its possession. Soil samples were collected at five sites for each of the three locations. At each sample site, vegetation and other debris were cleared and soil was collected to a depth of approximately 8 cm, placed in a 2-liter container, and marked with a sample identification number. At two of the sample sites, a second soil sample was collected to serve as a duplicate sample.

The soil samples were sieved into six size-fractions with physical diameters greater than 600 μm, 300 to 600 μm, 150 to 300 μm, 75 to 150 μm, 38 to 75 μm, and less than 38 μm. The 38 μm fraction was further separated by aerodynamic sizing into five size-fractions with aerodynamic diameters less than 1 μm, less than 2.5 μm, less than 5 μm, less than 10 μm, and less than 15 μm. From a dose perspective, particles less than 15 μm are considered potentially inhalable. No additional analysis was performed on the particle size-fraction between 15 μm and 38 μm. Quanterra Environmental Services, Richland Laboratory, analyzed the aerodynamically separated particle samples for plutonium, americium, and uranium using alpha spectroscopy. Uranium was found on single size-fractions in only four samples in low levels ranging from 1.12 to 3.72 pCi/g (0.06 to 0.11 pCi total activity) and was not considered in further analysis. UNLV's Department of Health Physics analyzed the sieved size-fractions for americium using gamma spectroscopy. Quality assurance/quality control included steps during sieving operations to minimum loss of material during shaking and during transfer from the sieves for weighing and analysis. For filter SSPSS operations and filter analysis, steps included the simultaneous collection of two <5 μm samples, random weighing of a control set of filters and with other calibration weights, leak tests and flow rate checks of the SSPSS equipment during operation, and a 5 to 10 minute “settling” period before filters were removed to help ensure that no particles remained in suspension.

For the aerodynamically separated size-fractions, analyzed activities for $^{239+240}$Pu ranged from 54 to 1,390 pCi/g for location one, from 450 to 9,000 pCi/g for location two, and from 83 to 1,600 pCi/g for location three. The analyzed activities for ^{241}Am ranged from 10 to 254 pCi/g for location one, from 14 to 1,490 pCi/g for location two, and from 14 to 220 pCi/g for location three. Several soil samples had more activity in the smaller size-fractions than in the larger ones, which suggests that radioactive particles may not have been uniformly distributed in the SSPSS suspension chamber during particle separation.

The average ratio of $^{239+240}$Pu to ^{241}Am for the aerodynamically separated samples analyzed by alpha spectroscopy for all size-fractions was 5.4 with a standard deviation of 1.5 and a range of 1.8 to 11.5, with the 95% confidence interval range being 5.1 to 5.8. The averages for the different size-ranges had slight but not significant differences. The ratio of 5.4 is three to four times lower than the ratio of $^{239+240}$Pu to ^{241}Am found for soil samples at safety tests on the TTR. The differences in the ratios for these isotopes compared to the TTR sites were also noted in research by the Nevada Applied Ecology Group at Project 57 (Gilbert et al., 1975).

The data indicate that there is a correlation between the aerodynamically separated particle size-fraction and its activity, although it is statistically more significant for plutonium than americium isotopes. The activities in the size-intervals for aerodynamic diameters <2.5 μm, from 2.5 to 5 μm, from 5 to 10 μm, and from 10 to 15 μm were determined from the mass and activities on filter samples. The mean activity of $^{239+240}$Pu on the aerodynamically separated samples increased as the particle size increased: 377 pCi/g for the <2.5 μm interval, 592 pCi/g for the 2.5 to 5 μm interval, 1211 pCi/g for 5 to 10 μm interval, and 1922 pCi/g for the 10 to 15 μm interval. Paired t-Tests show statistically significant increases in $^{239+240}$Pu activity between the <2.5 μm and 2.5-5 μm intervals (at the 3.4% level) and the 2.5-5 μm and 5-10 μm intervals (at the 1.3% level). The increase between the 5-10 μm and 10-15 μm interval had a significance level of 12.5% and would not be considered statistically significant. The means of the ^{241}Am activities in the <2.5 μm, from 2.5 to 5 μm, from 5 to 10 μm, and from 10 to 15 μm size-intervals were also computed. The ^{241}Am activities also tended to increase as the particle size increased, but no increase was statistically significant.

The activities of ^{241}Am in the sieved sample measured by gamma spectroscopy, particularly in the small size-intervals, are higher (range of 8.4 to 2630 pCi/g) than might be expected given the activities of ^{241}Am and $^{239+240}$Pu in the aerodynamically separated samples. If the same ratio of plutonium to americium holds as for the aerodynamically separated size-fractions, the plutonium activity of the sieved samples would be about five times the americium activity. The plutonium activity in the <38 μm size-range would then range from 800 to 13,000 pCi/g. The results of the gamma counting and subsequent calculations have been checked and determined to be correct to within the uncertainties of the measurement. A large amount of plutonium associated with particles having aerodynamic diameters greater than 15 μm (a diameter of about 3 μm for a plutonium particle or 5 μm for a plutonium oxide particle) and physical sizes less than 38 μm could result in the high activities in this size range.

In general, the relation among the sieved samples from the three locations is similar to that of the aerodynamically separated samples. The highest activities were found for location two; the activity at location three slightly exceeded that at location one. The individual soil samples with the highest alpha activities also had the highest gamma activities; the individual soil samples with the lowest alpha activities also had the lowest gamma activities. A few soil samples had one or more size-intervals with much higher activity than is found in other size-intervals for the sample, which may be indicative of the inhomogeneity of the mixture of radionuclides in some of the samples. However, in general, the results suggest than plutonium and americium are similarly distributed in the soil, both in the potentially inhalable size range and in larger size particle fractions.

The total amount of ^{241}Am in each sieved sample cannot be determined, because the sieved fractions were not weighed when the separations were done. Similarly, the total activity of the soil samples cannot be determined since plutonium analysis was not performed on the sieved fractions. The sieved fractions were not originally intended to be analyzed. The sieved fractions were analyzed only after a survey meter indicated relatively high activities in some of the sieved fractions. Previous work at the site (Papelis et al., 1996) indicated slightly more than 50% of the Pu activity is associated with particles less than 38 μm, but with considerable variability between samples. In any future, work with the SSPSS, the total weight soil sample, the weight of the part that

is sieved, and the weights of each sieved size-fraction should be determined. The principal benefit would be to more fully understand total activity distribution among all soil particle sizes.

5.0 REFERENCES

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